UNIVERSITAT ROVIRA I VIRGILI

DEVELOPMENT OF ENVIRONMENTAL TOOLS FOR THE MANAGEMENT OF SEWAGE SLUDGE ON AGRICULTURAL SOILS

Ana Carolina Passuello

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Ana Carolina Passuello

Development of environmental tools for the management of sewage sludge on agricultural soils

DOCTORAL THESIS

Supervised by

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Department of Chemical Engineering

Universitat Rovira i Virgili

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CERTIFY:

That the present study, entitled “Development of environmental tools for the management of sewage sludge on agricultural soils”, presented by Ana Carolina Passuello for the award of the degree of Doctor, has been carried out under our supervision at the Department of Chemical Engineering of this university, and that it fulfils all the requirements to be eligible for the European Doctorate Label.

Tarragona, December 20th 2010.

Marta Schuhmacher Ansuategui   Martí Nadal Lomas

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To my husband.

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Summary

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Sewage sludge is the main residue of wastewater treatment plants. This residue has proven an increase in its production, due to improvement of the wastewater collection and treatment systems. As any residue, sludge must be properly disposed to avoid risks to humans and ecosystems.

Several practices are applied for the management of SS. Among those, the application on agricultural soils is a preferred route for several countries, due to the possibility of recycling organic matter and nutrients to soil. Unfortunatelly, there are a few studies regarding the risks and benefits of this practice. In addition, there is a lack of proper regulation that considers all the factors related to its disposal on agricultural soils. The existing regulatory efforts concern metals concentrations in soils and sewage sludge. However, the levels of Persistent Organic Pollutants (POPs) in soils and sludge are not regulated so far.

As a consequence, stakeholders are not confident about the potential risks derived from sludge amendment, as risks determination present a high related uncertainty. The general population is concerned about the risks that may represent ingesting food produced, while farmers also want to keep the quality levels of their products. Finally, environmental agency is concerned about the potential consequences in terms of environmental pollution.

The objective of this thesis is developing tools to support the management of sewage sludge in agricultural soils. The thesis is divided in two main parts.

Part I (Chapters 3 to 5) presents the development, evaluation and integration of the fate, exposure and risk models. *Chapter 3* introduces a fugacity fate model. This model evaluates the trends for the concentrations of different groups of POPs for a time span of 20 years in agricultural soils amended with sewage sludge. In *Chapter 4*, the sensitivity analysis methods applied to the fugacity fate model are introduced. The evaluation results point out the most sensitive parameters of the fate model. *Chapter 5* regards fate, exposure and risk models integration and evaluation. Each of the integrated models is described and the results presented. Uncertainty and sensitivity analysis are performed through model integration.

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Part II (Chapters 6-9) describes the development of decision models. These decision models are integrated in Geographic Information Systems (GIS) to indicate the best agricultural areas to amend with sewage sludge. In *Chapter 6*, the problem of managing sewage sludge on agricultural soils is introduced. Also, the spatial multicriteria decision analysis framework is described and some methods for model integration, introduced. *Chapter 7* presents the application of the spatial multicriteria decision analysis framework to a case-study in the region of Catalonia (NE of Spain). The model is integrated and evaluated. The uncertainty of the decision models is evaluated in *Chapter 8*. The application of Bayesian networks to the decision model allows a broader evaluation of the same case study, as it considers the lack of knowledge related to model's development. Finally, *Chapter 9* summarises the most important conclusions from the results obtained in the framework described in this thesis.

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Resumen

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La producción de lodos procedentes de estaciones depuradoras de aguas residuales (EDAR) ha experimentado un aumento en los últimos años debido a las mejoras en los sistemas de recogida y tratamiento. Al igual que otros residuos, los lodos de depuradoras (LD) deben ser dispuestos adecuadamente para evitar riesgos en la población y los ecosistemas.

Para la gestión de lodos se pueden utilizar varias prácticas. Entre ellas, la aplicación en suelos agrícolas es la vía preferida por muchos países, ya que supone reciclar materia orgánica y aportar nutrientes al suelo. Desafortunadamente, el número de estudios relacionados con la evaluación del impacto de esta práctica es limitado. Además, hay una falta de legislación apropiada que tenga en cuenta todos los factores relacionados con la disposición en suelos agrícolas. Los esfuerzos legislativos existentes regulan los niveles de metales en suelos y lodos. Sin embargo, los niveles máximos de contaminantes orgánicos persistentes (COPs) en suelos y lodos aún no están contemplados por la ley.

Como resultado, los grupos de interés (stakeholders) no están seguros acerca de los potenciales riesgos derivados de la utilización de los lodos como fertilizante agrícola, ya que la determinación de los riesgos presenta elevada incertidumbre. Mientras que la población general se preocupa por los riesgos de ingerir alimentos producidos en estas zonas, los agricultores quieren mantener la calidad de su producto y, por último, las agencias medioambientales tienen por objeto evitar una posible contaminación del medio.

El objetivo de esta tesis es desarrollar herramientas de apoyo a la gestión de los lodos de depuradora en suelos agrícolas. Estas herramientas deben tener en cuenta las propiedades ambientales relacionadas con la gestión de los lodos. La tesis se divide en dos partes.

Parte I (capítulos 3-5) presenta el desarrollo, evaluación e integración de los modelos de transporte, exposición y riesgo. El *Capítulo 3* presenta el modelo de transporte basado en el concepto de fugacidad. Este modelo evalúa las tendencias de las concentraciones de COPs en suelos abonados con lodos, para un período de 20 años.

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En el *Capítulo 4*, se introducen los métodos de análisis de sensibilidad aplicados al modelo de fugacidad. Los resultados de la evaluación indican los parámetros más sensibles del modelo de transporte. El *Capítulo 5* presenta la integración y evaluación de modelos de transporte, exposición y riesgo. Se describe cada uno de los modelos integrados y se presentan los resultados. Los análisis de incertidumbre y sensibilidad se realizan durante la integración de los modelos.

La *Parte II* (Capítulos 6-9) describe el desarrollo de modelos de toma de decisiones. Estos modelos están integrados en Sistemas de Información Geográfica (SIG) para indicar las mejores zonas agrícolas para ser fertilizadas con lodos. En el *Capítulo 6*, se introduce el problema de la gestión de los lodos en suelos agrícolas. Además, el análisis espacial de la toma de decisión multicriterio se estructura y se describen algunos métodos de integración. El *Capítulo 7* presenta la aplicación del análisis espacial de decisión multicriterio en un caso práctico para Cataluña. La incertidumbre relacionada con los modelos de decisión se evalúa en el *Capítulo 8*. La aplicación de redes Bayesianas al modelo de decisión permite una evaluación más amplia del mismo caso, teniendo en cuenta la falta de conocimiento relacionada con el desarrollo del modelo. Por último, en el *Capítulo 9* se resumen las conclusiones más importantes a través de los resultados obtenidos en el ámbito de esta tesis.

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Resumo

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A produção de lodos provenientes de Estações de Tratamento de Esgoto (ETE) experimentou um incremento nos últimos anos, devido a melhorias nos sistemas de captação e tratamento. Como qualquer resíduo, estes lodos devem ser dispostos apropriadamente para evitar riscos à população e aos ecossistemas.

Várias práticas podem ser utilizadas para a gestão de lodos. Entre elas, a aplicação em solos agrícolas é a via preferida para muitos países, devido à possibilidade de reciclar matéria orgânica e nutrientes ao solo. Infelizmente, o número de estudos relativos a esta prática é limitado. Ainda, existe uma falta de legislação apropriada que considere todos os fatores relacionados à disposição em solos agrícolas. As iniciativas legais existentes regulam níveis de metais em solos e lodos. Porém, os níveis máximos de Contaminantes Orgânicos Persistentes (COPs) em solos e lodos ainda não estão contemplados na lei.

Em conseqüência, os grupos interessados (stakeholders) não estão seguros a respeito dos riscos potenciais derivados do uso de lodos como fertilizante agrícola, já que a determinação destes riscos apresenta elevada incerteza. Enquanto a população em geral preocupa-se com os riscos relacionados à ingestão de alimentos produzidos, os agricultores desejam manter os níveis de qualidade de seu produto, e finalmente, a Agência Ambiental pretende prevenir conseqüências potenciais em termos de poluição ambiental.

O objetivo desta tese é desenvolver ferramentas para apoiar a gestão de lodos de ETE em solos agrícolas. A tese está dividida em duas partes.

A *Parte I* (Capítulos 3 a 5) apresenta o desenvolvimento, avaliação e integração dos modelos de transporte, exposição e risco. O *Capítulo 3* introduz o modelo de transporte baseado no conceito de fugacidade. Este modelo avalia as tendências para as concentrações de diferentes grupos de COPs em solos adubados com lodos, para um período de 20 anos. No *Capítulo 4*, os métodos de análise de sensibilidade aplicados ao modelo de fugacidade são introduzidos. Os resultados desta avaliação indicam os parâmetros mais sensíveis do modelo de transporte. O *Capítulo 5* apresenta a integração e avaliação dos modelos de transporte, exposição e risco. Cada um dos

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modelos integrados é descrito e os resultados apresentados. Análises de incerteza e sensibilidade são executadas durante a integração dos modelos.

A *Parte II* descreve o desenvolvimento de modelos de tomada de decisão. Estes modelos são integrados em Sistemas de Informação Geográfica (SIG) para indicar as melhores áreas agrícolas para serem adubados com lodos. No *Capítulo 6*, o problema de gerir lodos em solos agrícolas é introduzido. Ainda, a análise espacial de tomada de decisão multicritério é estruturada e alguns métodos de integração descritos. O *Capítulo 7* apresenta a aplicação da análise espacial de tomada de decisão multicritério em um estudo de caso para a região da Catalunha (NE da Espanha). Este modelo é integrado e avaliado.

A incerteza relacionada aos modelos de decisão é avaliada no *Capítulo 8*. A aplicação de redes Bayesianas ao modelo de decisão permite uma avaliação mais ampla do mesmo estudo de caso, por considerar a falta de conhecimento relacionada ao desenvolvimento de modelos.

Finalmente, o *Capítulo 9* resume as conclusões mais importantes dos resultados obtidos no âmbito desta tese.

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Abbreviations

A	Arithmetic mean operator
ACA	Catalan Water Agency (Agència Catalana de l'Aigua)
AHP	Analytic Hierarchy Process
BNs	Bayesian networks
CA	Partial conjunction operator
$CaCO_3$	Carbonates
Cd	Cadmium
CEC	Council of the European Communities
CPT	Conditional probability table
Cr	Chromium
Cu	Copper
C_xH_y	Hydrocarbon
DA	Partial disjunction operator
DAG	Directed Acyclic Graph
dw	Dry weight
EC	European Commission
GCD	Generalized conjunction/disjunction
GIS	Geographic Information Systems
GSA	Global sensitivity analysis

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GW	Groundwater
HCl	Hydrogen chloride
HF	Hydrogen fluoride
Hg	Mercury
HLs_p	Half life on soil
H_y	Henry constant
K_{MW}	Mineral matter-water partition coefficient
K_{OC}	Sorption partition coefficient
k_{OW}	Octanol/Water Partition Coefficient
LHS	Latin Hypercube Sampling
LOAEL	Lowest-observed adverse-effect level
Log	Logarithm base 10
LSP	Logic Scoring of Preference
MAPA	Ministry of Agriculture, Fisheries and Food of Spain
	(Ministerio de Agricultura, Pesca y Alimentación de España)
MAUT/MAVT	Multi-Attribute Utility/Value Theory
MCDA	Multicriteria decision analysis
MMA	Ministry of the Environment and Rural and Marine Affairs of Spain
	(Ministerio de Medio Ambiente y Medio Rural y Marino de España)
MS	Microsoft

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N	Nitrogen
N_2O	Nitrous oxide
NE	Northeast
Ni	Nickel
NOAEL	No-observed-adverse-effect level
NO_x	Nitrogen oxide
OCDD	Octachlorodibenzodioxin
OCDF	Octachlorodibenzofuran
ODE	Ordinary differential equation
OM	Organic matter
OWA	Ordered Weighted Averaging
PAHs	Polycyclic aromatic hydrocarbons
Pb	Lead
PCBs	Polychlorinated biphenyls
PCC	Partial Correlation Coefficient
PCDD/Fs	Polychlorinated dibenzo-*p*-dioxins and dibenzofurans
pdf	Probability distribution function
PeCDF	Pentachlorodibenzofuran
POPs	Persistent Organic Pollutants
PRA	Probabilistic risk assessment

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R^2	Coefficient of determination
RD	Royal Decree
RMSE	Root mean square error
SA	Sensitivity Analysis
SD	Standard deviation
SF	Slope factor
SMCA	Spatial Multicriteria Decision Analysis
SO_2	Sulfur dioxide
SRC	Standardized Regression Coefficient
SS	Sewage sludge
TCDD	Tetrachlorodibenzodioxin
USEPA	United States Environmental Protection Agency
VOC	Volatile organic compounds
WWTP	Wastewater treatment plants
Zn	Zinc

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Equations symbols

AFI_g	Gastrointestinal absorption (human)
A_L	Leaf area
AT	Averaging time of exposure
B_A	Molecular diffusivity in pore air
Ba	Biotransfer factor in beef and milk
B_W	Molecular diffusivity in pore water
BW	Body weight
CDI	Chronic daily intake of the chemical
C_{jp}	Concentration of the chemical *p* in compartment *j*
$C_{gas,p}$	Concentration of the chemical p in the gas phase
$C_{L,p}$	Concentration in of contaminant *p* in leaves
$C_{part,p}$	Concentration of the chemical *p* in air particle phase
$C_{S,p}$	Concentration of the chemical *p* in soil
CR	Cancer risk
CR_s	Soil ingestion rate (farmer)
CR_{beef}	Consumption rate of beef (human)
CR_{milk}	Consumption rate of milk (human)
CR_{veg}	Consumption rate of vegetables (human)
CSF	Cancer slope factor

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$diff_i$	Diffusion distance in layer *i*
D_{ip}	Input transfers in layer *i* for pollutant *p*
$D_{kp}f_{kp}$	Flow rates from layer *k* to layer *i* for pollutant *p*
$D_{ip}f_{ip}$	Total outputs from layer *i* for pollutant *p*
D_{ikAp}	Diffusion in pore air of contaminant *p* from layer *i* to the layer *k*
D_{ikBp}	Sorbed phase transport (due to bioturbation) of contaminant *p* from layer *i* to the layer *k*
D_{ikWp}	Diffusion in pore water of contaminant *p* from layer *i* to the layer *k*
D_{iLp}	Leaching (L) of contaminant *p* from layer *i* to the layer below
D_{iRp}	Degrading reactions of contaminant p in layer *i*
D_{iVp}	Volatilization of contaminant *p* in layer *i*
Ei	Preference of each criterion *i*, in the LSP method
ED	Exposure duration
EF	Exposure frequency
ET	Exposure time
EU	European Union
FI_{cattle}	Fraction of food intake by the animal from the contaminated area
FI_{human}	Fraction of food intake by humans from the contaminated area
f_{ip}	Fugacity of pollutant *p* fugacity for the layer *i*
g	Conductance for diffusive transfer between leaves and air

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HQ	Non-carcinogenic risk or hazard quotient
I_i	Input rate in layer *i* for pollutant *p*
IR	Ingestion rate
IUR	Inhalation Unit Risk
k	First order degradation rate (in plant leaves)
K_{LA}	Partition coefficient between leaves and air
K_{SA}	Soil-air partition coefficient
L	Leaching rate
M	Maximum level of metals in soils allowed by the legislation
M_{ip}	Molar concentration of pollutant *p* in layer *i*
M_{p0}	Total mole amount of *p* at the initial time
$M_{L,p}$	Molar concentration of *p* in leaf
M_{pt}	Total mole amount of pollutant *p* at time *t*
MP	Most probable
MW_p	Molecular weight of pollutant *p*
N	Number of evaluated pixels or sample size
OC_{OM}	Mass fraction of OM in soil OC
OC_{SSi}	Mass fraction of OC in soil solids in layer *i*
P	Preference value
p	Threshold of strict preference

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PCC	Partial correlation coefficient
PE	Percent of error
P_{High}	Probability of having a high suitability
P_{Low}	Probability of having a low suitability
P_{Medium}	Probability of having a medium suitability
$P_{Very\ high}$	Probability of having a very high suitability
P(Y)	Normalization term
$P(Y\|\theta)$	Distribution of known quantities, Y, given unknown quantities, θ, or likelihood
$P(\theta)$	Prior distribution of unknown quantities θ
$P(\theta\|Y)$	Posterior distribution of unknown quantities, θ, given known quantities, Y
q	Threshold of indifference
Q_L	Transpiration stream on leaves
Qp_{beef}	Quantity of plant eaten by beef cattle
Qp_{milk}	Quantity of plant eaten by dairy cattle
Qs_{beef}	Soil consumption rate for beef cattle
Qs_{milk}	Soil consumption rate for dairy cattle
r	Exponent used to adjust logic properties of LSP aggregation function
$RemRate_{pt}$	Removal rate at different time-spans

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RfC_i	Inhalation Reference Concentration
RfD	Reference dose
RfD_o	Oral Reference Dose
S	Sensitivity coefficient
SF_o	Slope Factor Oral
S_i	Suitability of the scenario map
S_{it}	Suitability of the original map
t	Time
TSCF	Transpiration stream concentration factor
V_B	Bioturbation velocity
V_i	Volume of soil layer *i*
V_{ij}	Volume of compartment *j* in layer *i*
V_L	Equivalent volume of above ground plant tissues
$v_{W,L}$	Volume fraction of water on leaves
Wi	Weight associated to each criterion *i* (LSP)
$\bar{x}$	Inputs mean value
x_l	Input value for run *l*
$\bar{y}$	Outputs mean value
Y_B	Air boundary layer thickness

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y_f^{all}	Value of the y variable estimated using a regression that considers all input variables.
$y_f^{\sim xh}$	Estimation of the y variable value using a regression that does not include input variable *xh*
y_l	Output value for run *l*
Z_{ijp}	Fugacity capacity in compartment *j* in layer *i* for pollutant *p*
Z_{ip}	Fugacity capacity or bulk Z value at the i-th layer for pollutant *p*
β	Standardised regression coefficient
ρ	Leaf density
σ	Standard deviation
u_{dep}	Deposition velocity of particles on leaves
◊	Generalised conjunction disjunction function

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Chapter 1

Introductory notes

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1. Introduction

Sewage sludge (SS) is the main residue generated at wastewater treatment plants (WWTP) in consequence of solids removal in various parts of the treatment system. It includes the sludge originated in treatment plants of domestic wastewater that is occasionally mixed with industrial wastewater and/or run-off rain water. Sewage sludge properties depend basically on the wastewater pollution load and technical characteristics of the treatment plant. Due to water treatment, the pollution present in water is concentrated and also some chemicals are transformed during the treatment process (Katsoyiannis and Samara, 2005).

According to Schowanek et al. (2004), "the organic fraction of sludge is a mixture of fats, proteins, carbohydrates, lignin, amino acids, sugars, celluloses, humic material, and fatty acids. Live and dead micro-organisms constitute a large proportion of the organic material and provide a large surface area for sorption of lipophilic organic contaminants in the sludge. The properties of sludge are dependent upon its origin and treatment type. It differs in its physical (processability and handlability), chemical (presence of nutrients and contaminants), and biological parameters (microbial activity and presence of pathogens). The characteristics of the sludge in terms of contaminant loading is dependent upon the original pollution loading of the sewage (i.e., domestic, industrial, and mixed) and the type of wastewater treatment the sewage and sludge have received. In many cases, some form of post-treatment is applied to the sewage sludge (e.g., anaerobic digestion or aerobic composting), and this can have a major impact on residual contaminant levels in case these are biodegradable."

During the last few decades, the increase of sludge production, as a consequence of industrial development, population increase and amplification of treatment services, has become an environmental problem for many countries. Sludge can be recycled or disposed using several routes. The most widely available options in the European Union are the agriculture utilization, the waste disposal sites, and the incineration (Aubain et al., 2001). Each of these disposing routes has related impacts and costs, and the choice of one alternative depends mainly on local characteristics, such as

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economic, social, legal and geographic characteristics of the region where SS is produced. The main advantages and disadvantages of each method are described in Table 1.1.

Table 1.1. Advantages and disadvantages of the most used managing practices. Source: Aubain et al. (2001) and Fytili and Zabaniotou (2008).

	Advantages	Disadvantages
Agricultural reuse	Fertilizing benefits (nitrogen, phosphorous, potassium, organic matter). Yield improvement. Fertiliser substitution.	Contaminants, such as heavy metals and persistent organic pollutants, may enter the food chain. Leaching, runoff and volatilization may transfer contaminants to air and water. Storage: SS can only be applied once or twice a year. Public acceptance.
Incineration	Reduction of sludge volume. Destruction of some organic compounds. Energy recovering, due to the calorific value of sewage sludge. Minimization of odour generation.	Generation of ashes (30% of the solids remain as ashes), that are highly toxic due to its metal's content and must be properly disposed. Air emissions of contaminants generated in the combustion, such as heavy metals, dioxins and furans, NO_x, N_2O, SO_2, HCl, HF and C_xH_y. Impacts on human health, ecosystems and climate change. Dried sludge needed. Water emissions due to wastewater production. Adverse health effects and ecotoxicity.
Landfilling	Co-generation of energy when the gas is collected.	Generation of gas: methane and carbon dioxide. Reported traces of VOC emissions (halogenated hydrocarbons and hydrocarbons). Generation of leachate that contains heavy metals, organic compounds (chlorinated organics, phenol, benzene, pesticides) and micro-organisms. Soil and water contamination, impacts to humans and ecosystems. Availability of disposal sites.

Among these routes, agricultural reuse is the preferred in several European countries (Aubain et al., 2001). In Spain, the annual SS generation raised more than 64%

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between the years 1997 and 2006 (MMA, 2008). The recycling of sludge to agriculture could handle the increasing quantities of sludge produced over the last years, representing 66% of the total disposal for the year 2006 (MMA, 2008).

According to the Spanish legislation, SS must be treated prior to application on land (MAPA, 1990). The selection of the treatment is related to the final application of sludge. Aubain et al. (2001) describes five groups of treatment processes: conditioning, thickening, dewatering, stabilization and/or disinfection, and thermal drying. Among those, stabilization and disinfection are the most attractive to treat sludge prior to its application on land. Biological stabilization and disinfection processes include anaerobic and aerobic digestion and composting. These treatments generally reduce the odour generation as well as the pathogen content of sludge. In addition, composted sludge presents a higher agricultural value, with a humus-like aspect, being easier to handle due to its low humidity and more accepted by farmers' community. Dewatering and thermal drying may be also used to reduce the water content of sludge, reducing also the transport costs. Once treated, the sludge is also referred as biosolids.

With appropriate use, the application of sewage sludge on agricultural fields has benefits to soils and crops (Schowanek et al., 2004). This amending practice improves soil fertility, reduces the use of fertilizers, and is a relatively inexpensive solution. It also improves the soil structure by the addition of organic matter (OM) to the soil matrix and recirculation of nutrients through soil. When compared to other types of fertilizer, such as urban compost and animal manure, sewage sludge, especially the composted, has higher OM concentration.

Amending soils with sludge may also have other positive effects. Chambers et al. (2003) observed a trend towards high water-infiltration rates and a decrease of surface runoff and erosion potential in amended soils. The authors also noticed an increased soil porosity and strength, improving the potential for air and water movement through the topsoil.

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Sludge content of nitrogen is also an important factor for the use of this residue as an organic amendment. Sludge value as a fertilizer is determined by the capacity of the organic nitrogen to be mineralized, as nitrogen is mostly found under organic form in sludge. The nitrogen availability is influenced by the type of sludge, temperature, humidity, pH and texture of the soil, as well as the application conditions (Aubain et al., 2001). The content of N in sludge may vary depending on the sludge treatment type, operational and storage conditions (Mantovi et al., 2005).

1.1 Health risks regarding soil amendment with sewage sludge

Although the application of sewage sludge on soils represents a number of benefits to the soil structure, there are health risks related to this practice. As any residue, SS may present important concentrations of some pollutants.

Maximum levels of metals on sludge and soils are regulated by the Council Directive 86/278/EEC (CEC, 1986). Aubain et al. (2001) presented a review of sludge characterization for European countries. They observed metal levels lower than those defined by the Directive. Metals are naturally present in different concentrations in background soils, among others, due to local geological characteristics of the field. They may be also originated from anthropogenic sources such as fertilizers, animal manure and sludge application. A wide range of metal concentrations in soil are reported in the literature (Fytili and Zabaniotou, 2008; Nadal et al., 2004a, 2007).

An extensive and diverse range of organic compounds are known to exist in sewage sludge (Katsoyiannis and Samara, 2005), many of which become transferred to sludge-amended agricultural soils (Wild and Jones, 1992). The presence of Persistent Organic Pollutants (POPs) in sludge has been reported in many investigations (Eljarrat et al., 2003, 2008; Harrison et al., 2006). POPs are toxic and persistent chemicals, characterized by their bioaccumulation potential and long-range transport capacity. These pollutants move through different environmental compartments. Within these fate processes, POP concentration dynamically changes over time and leads to accumulation in the food chain. As a final step, these contaminants may be transferred

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to human beings. POPs have been also found in soils from industrial, urban and even rural zones (Nadal et al., 2004b, 2007, 2009a; Wyrzykowska et al., 2007).

The European Community is aware of the necessity of including organic contaminants in the regulations. So far, the 3rd draft of the working document in sludge (CEC, 2000) defines limit values for concentrations of organic compounds in sludge intended for use on land. However, these levels are classified according to groups of contaminants, not considering the persistence, accumulation and toxicity properties of each congener. There is a lack of studies regarding POP fate due to soils amendment, in which long-range transport and human exposure to these contaminants are considered. In order to determine the safe levels of POPs in soils and food, environmental models must be performed.

Environmental models are tools to predict pollutants distribution on the different media and evaluate the risk for the population, through exposure and risk assessment models. Among them, fugacity-based models have been largely used to determine the environmental distribution of POPs (Mackay and Fraser, 2000, Czub and McLachlan 2004a, Wania et al. 2006). Fugacity is a convenient quantity to describe mathematically the rates at which chemicals diffuse, or are transported, between phases (Mackay, 2001), since the transfer rates are proportional to the fugacity difference between sources and destination compartments.

The case of soil amendment is a complex one, as a point (on time) application is performed (once or twice a year) at a large area, which characterizes diffuse pollution. In addition, the system is open (equilibrium is not reached) and non-steady state (Hughes et al., 2005). The high persistence of the contaminants only increases the system complexity and adds high uncertainty on system parameters estimation. In consequence, a crucial point of this study is the design of a multimedia POPs fate and exposure model, for the case of SS amendment on agricultural soils.

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1.2 Sewage sludge management on agricultural soils

Environmental management has become an issue of increased interest in the last few years. This is mainly a consequence of the complexity of the environmental systems, added to the existence of stakeholders with conflictive objectives. The case of sewage sludge management is not different. SS production has recently increased due to population growth and improved collecting and treatment systems. Managing the increased production of this residue has become a challenge for several countries.

Nevertheless, the current management practices do not reflect all the factors that should be taken into account for the application of SS on agricultural areas. There are few legislative efforts to regulate this practice. In addition, the population is not well informed about the related risks and benefits. As a consequence, the decisions are not as confident as they should be, and stakeholders may find that their interests are not reached, leading to a low acceptance of the practice. A way of avoiding that is through participatory planning, which can enhance the sustainability and public acceptance of overall management (Holzkaemper et al., 2009), as it is performed with the support of all interested parts. The engagement of population, farmers, companies and environmental agencies is crucial to define the best management options.

Amajirionwu et al. (2008) conducted a survey with Irish people to determine their main concern regarding sludge amendment. They found that the most serious concerns are the damage to human health, the need for protection of clean air, water and soil, the damage to animal health, the possible contamination of biosolids and the damage to plants and crops, in this order. Other problems regard the proximity of the population to the storage and processing sites, the objectionable smells, fear of loss of property and land value, among others.

Farmers are mainly concerned about food quality, as they must respect quality standards to avoid reducing their market share. For them, SS application may be a profitable practice due to the saving of fertilizers and management costs.

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The environmental agency must assure that good environmental levels are maintained. For that, soil and landscape characteristics must be evaluated for each case to avoid environmental contamination. Finally, companies are hired by the environmental agency to distribute SS in agricultural areas. These companies are mainly interested on profit as long as they accomplish the limited existing legislation.

Deal with conflicting interests is a complex issue that involves consideration of multiple objectives (e.g., environmental, ecological) relevant to evaluating and selecting among the management alternatives. For that, the development of a tool that deals with the integration of these objectives is critical.

In this specific case, the study of the availability of land is of interest, because while SS production increases, the availability of land is limited. The evaluation of the suitability of agricultural areas, considering environmental and ecological criteria is needed to guarantee the future sustainability of these agricultural areas that use sludge as an organic amendment. The development of decision tools, coupled with GIS, allow more confident decisions through the evaluation of the territory, indicating the different suitability levels of the existing agricultural areas.

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2 Hypothesis

Sewage sludge is a residue that must be properly disposed to protect the environment. The amendment of soils with sludge is the most applied option nowadays, especially in Spain. This option has absorbed more than 65% of the total sludge production for this country, for the last few years. With appropriate use, this practice has benefits to soils and crops. However, as any residue, sludge may also represent a contamination load. Levels of metals are controlled by legislation, but there is not any legal effort regarding POPs.

POPs are man-made chemicals characterized by a high persistence and accumulation potential with important toxicity values. These contaminants move in the environmental matrices and may reach the food chain. Due to its novelty, there is a lack of studies regarding human exposure to persistent organic pollutants because of the use of sludge as an organic amendment.

In addition, there is a need of inserting stakeholders' expectations in SS management. An improved acceptance of the practice could be proved if the interests of all stakeholders are taken into account. A broader understanding of the managing problem is needed to promote the confidence on the practice.

In conclusion, it is hypothesized that amending soils with sewage sludge could represent a risk to human health due to the presence of POPs in the sludge matrix. Likewise, there are some agricultural areas that might be more indicated for receiving sewage sludge as an organic amendment than others. The classification of these areas depends on local characteristics of the fields.

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3 Objectives and Thesis Outline

3.1 General objective

To develop environmental tools to assure the best possible management of sewage sludge on agricultural soils. These tools must consider environmental properties related to sewage sludge management.

3.2 Specific objectives

- To determine the fate of POPs in soils for a time span of 30 years.
- To assess the transference of POPs from soil to food: crops, cattle meat and milk.
- To evaluate human exposure to POPs as a consequence of SS amendment on agricultural soils and calculate the related human health risks.
- To determine the most important factors in POPs fate, human exposure and health risk models.
- To develop decision tools for land classification to determine the best areas to amend with SS.
- To determine the most important factors on land suitability.
- To represent the uncertainty related to each developed model.

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3.3 Thesis outline

In order to reach the defined objectives, some steps must be followed. The first two steps concern fate, exposure and human health risk evaluations due to the presence of POPs is sewage sludge applied to land (Part I). The last two steps regard decision making, with the selection of the best agricultural fields to amend with sludge (Part II). The thesis general scope is presented in Figure 1.1.

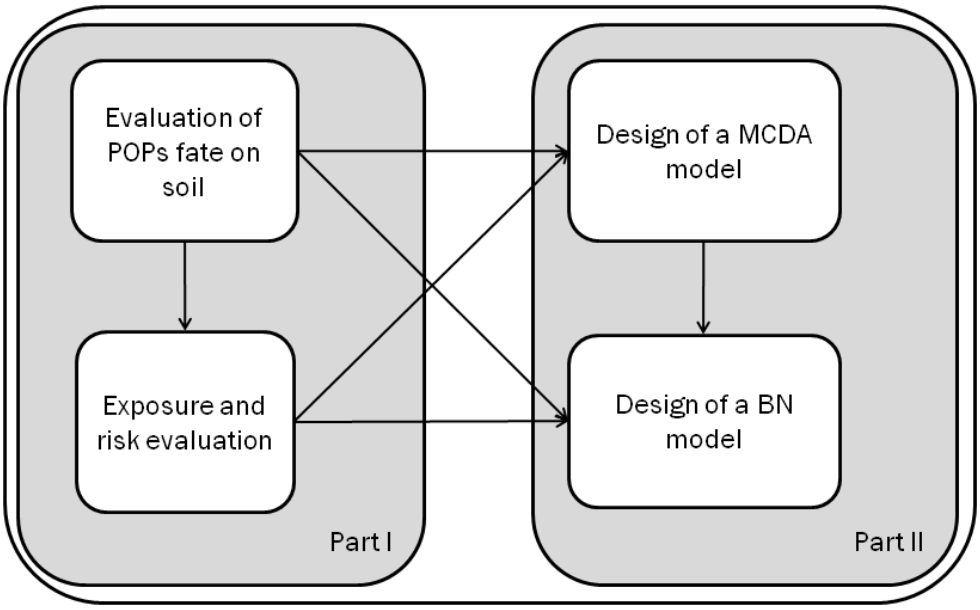

Figure 1.1. Thesis general scope

The thesis is structured as follows. After a short introduction about the problem of disposing sludge on agricultural soils in *Chapter 1*, *Chapter 2* introduces all the methods applied into this thesis.

Chapters 3 to 8 present the implementation of the methods to the case-study. Each chapter describes the development and application of a specific environmental tool. As a consequence, each chapter is defined as an independent unit and is totally understandable without the need of referring to the other chapters. To promote a better reading and comprehension, the thesis is divided in two parts (Part I and II).

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Part I (Chapters 3 - 5) presents the development, evaluation and integration of the fate, exposure and risk models. *Chapter 3* presents a fugacity fate model to evaluate the long range transport of POPs in agricultural soils amended with sewage sludge. The chapter presents the trends for the concentrations of different groups of POPs for a time span of 20 years.

Due to the complexity of the model developed, a previous evaluation must be performed. *Chapter 4* presents the sensitivity analysis of the method described in *Chapter 3*. Two methods are presented: the standardized regression coefficients and the partial correlation coefficients. These methods indicate the most sensitive parameters of the fate model.

In *Chapter 5*, several models are presented and integrated. The results of the fate model are used as an input for these models. The plant model estimates the concentration on leaves for two types of plants, lettuce and grass, considering three uptake processes: deposition, diffusion and root transfer. The food chain model considers the ingestion of grass and soil by cattle. Finally, the human exposure and health risk model evaluates two pathways (ingestion and inhalation) for two scenarios (occupational and non-occupational). The ingestion pathway considers the ingestion of soil, vegetables, meat and milk, while the inhalation pathway considers particle inhalation. Uncertainty and sensitivity analysis are performed through model integration.

Part II (Chapters 6 - 8) describes the development of two decision models to indicate the best agricultural areas to amend with sewage sludge. Due to the complexity of the decision problem, *Chapter 6* is entirely dedicated to the description of the problem and the development of a spatial multicriteria decision analysis to deal with this problem. The involved stakeholders are described and a criteria structure is suggested. The spatial multicriteria decision analysis framework is presented, while some methods for model integration are introduced.

Then, in *Chapter 7*, the described framework is applied to a case-study in the region of Catalonia (NE of Spain). After presenting the results of the integration in GIS, the model

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is evaluated based on two methods. Global Sensitivity Analysis is performed to select the most sensitive criteria. Then, the exploratory method is applied to assess the error related to the elimination of each one of the most sensitive criteria. Together, the evaluations allow defining model sensitivity to specific case-study inputs.

In *Chapter 8*, model uncertainty is inserted in the evaluation through the application of Bayesian networks (BNs). The method gives a broader evaluation of the same case-study, as it considers the lack of knowledge related to model development. The results, presented in GIS, give a probability of reaching each one of the defined suitability states. The results representation through conditional probability tables eases the identification of the sources of uncertainty for each evaluated pixel.

Finally, in *Chapter 9* the most important results are summarized and the conclusions of this thesis are presented.

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Chapter 2

Methods overview

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1 Environmental models

Environmental models are tools that help to predict and evaluate natural phenomena. Among these, fugacity models (item 1.1) allow predicting the way chemicals diffuse and move between environmental compartments, while exposure and risk models (item 1.2) evaluate the exposure levels and the likelihood of developing diseases based on human exposure. These models are crucial to guarantee that human interventions on the environment are performed at safe levels.

1.1 Fugacity models

Fugacity models have been largely applied in the field of environmental modelling to evaluate the fate of organic contaminants. Webster and Mackay (2007) report that: “Fugacity was introduced by G.N. Lewis (1901) as a criterion of equilibrium. It is similar to chemical potential, but unlike chemical potential, it is proportional to concentration, at least for most environmental conditions.” The application of the fugacity concept to environmental models was introduced and fully described by Mackay (2001).

Fugacity means a tendency to escape or pass away. It has units of pressure (Pa) and may be understood as a partial pressure exert by a chemical to escape from one phase or compartment to another. When equilibrium is achieved a chemical has the same fugacity in all phases, although concentrations may differ.

However, when the fugacity of one matrix exceeds the fugacity of other the contaminant moves from the higher to the lower fugacity medium, until the equilibrium is reached. Due to this fact, fugacity is a convenient quantity for describing mathematically the rates at which chemicals diffuse, or are transported, between phases. An example is the volatilization of pesticides from soil to air. The transfer rate can be expressed as being driven by, or proportional to, the fugacity difference that exists between the source and destinations phases (Mackay, 2001).

The advantage of fugacity is that for a compartment such as a lake containing water, if suspended solids and biota are at equilibrium, a single fugacity applies thus a single

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mass balance equation is written. The number of mass balance equations equals the number of fugacities.

Mackay et al. (2001) classifies fugacity models according to their increasing complexity.

Level I models show the relative equilibrium partitioning of a conserved (i.e. non-reacting) chemical in a multimedia setting. They assume equilibrium and steady-state to apply in a closed system, and no account is taken of reactivity. For the Level I calculation, both soil and sediment are treated as simple solid phases and the presence of air or water in the pores of these phases is ignored (Mackay et al., 2000).

Level II models include degrading reactions and advective loss but they assume that all media are at equilibrium, so only one fugacity and one mass balance equation apply. No intermedia mass exchange rates are deduced, i.e. all resistances are neglected, and equilibrium is achieved instantaneously. Therefore, chemicals immediately establish equilibrium when introduced to the system and the mode-of-entry is irrelevant.

Level III models assume steady-state, i.e., conditions are constant with time but compartments are not at equilibrium and different fugacities apply to each medium. In consequence, intermedia mass transfer resistances are not zero and the mass fractions depend on the mode-of-entry. The overall persistence is thus a function of how the mass enters the system (Mackay et al., 2001).

Level IV models are dynamic or unsteady-state in nature. The compartments are not in equilibrium and the inputs are constantly changing. Different fugacities and transfer rates are applied. These models are most often used to determine how long it will take for concentrations to change as a result of changing rates of emission. This is the case of sewage sludge amendment on agricultural soils.

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1.1.1 Basis for Level IV model calculation

The soil model developed in this thesis considers 4 compartments – pore air (A), pore water (W), organic matter (OM) and mineral matter (MM) - in a two layer soil (top and bottom). The model input parameters are the following: sludge characteristics and application dose, soils characteristics (density, texture, organic matter content, transport parameters) and pollutant properties (solubility, k_{OW}, K_{OC}, vapor pressure, Henry constant and half-life in soil).

For a given chemical *p*, the fugacity of a layer *i* (f_{ip}, Pa) and the concentration a compartment *j* of this layer (C_{ijp}, mol m^{-3}) are related according to Equation 2.1:

$$C_{ijp} = Z_{ijp} f_{ip} \tag{2.1}$$

Where Z_{ijp} is the fugacity capacity in compartment *i* (mol m^{-3} Pa^{-1}). According to Webster and Mackay (2007), Z_{ijp} expresses the capacity of a phase, or environmental medium, for a given chemical. Z_{ijp} values are large when the chemical is readily soluble in a phase, i.e., the phase can absorb a large quantity of the chemical. A low Z_{ijp} value indicates that the phase can accept only a small quantity of chemical, i.e., the chemical is “less-soluble” in the phase.

The fugacity (Equation 2.2) of a contaminant on each layer *i* is proportional to the molar concentration (M_{ip}) of the contaminant and inversely proportional to the Z_{ijp} value and the compartment volume (V_{ij}, m^3):

$$f_{ip} = \frac{M_{ip}}{V_{ij} Z_{ijp}} \tag{2.2}$$

Finally, the mass balance differential equation for a compartment *j* is described by the equation (2.3):

$$V_{ij} Z_{ijp} \frac{df_{ip}}{dt} = I_{jp} + \sum \left(D_{kp} f_{kp} \right) - \sum \left(D_{ip} f_{ip} \right) \tag{2.3}$$

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Where t is the time (h), I_{jp} is the input rate (mol h^{-1}), D_{kp} represent the input transfers from layer k (mol Pa^{-1}·h^{-1}) and $D_{ip}f_{ip}$ is the total output (mol h^{-1}). For the complete description of these transfer processes, please refer to Webster and Mackay (2007).

1.2 Risk assessment

Risk assessment may be defined as a tool to estimate the likelihood of an adverse effect occurring, and the probability and severity of adverse consequences from an exposure of potential receptors. In general terms, risk assessment is performed through 4 steps: hazard identification, exposure assessment, dose-response assessment and risk characterization (Figure 2.1).

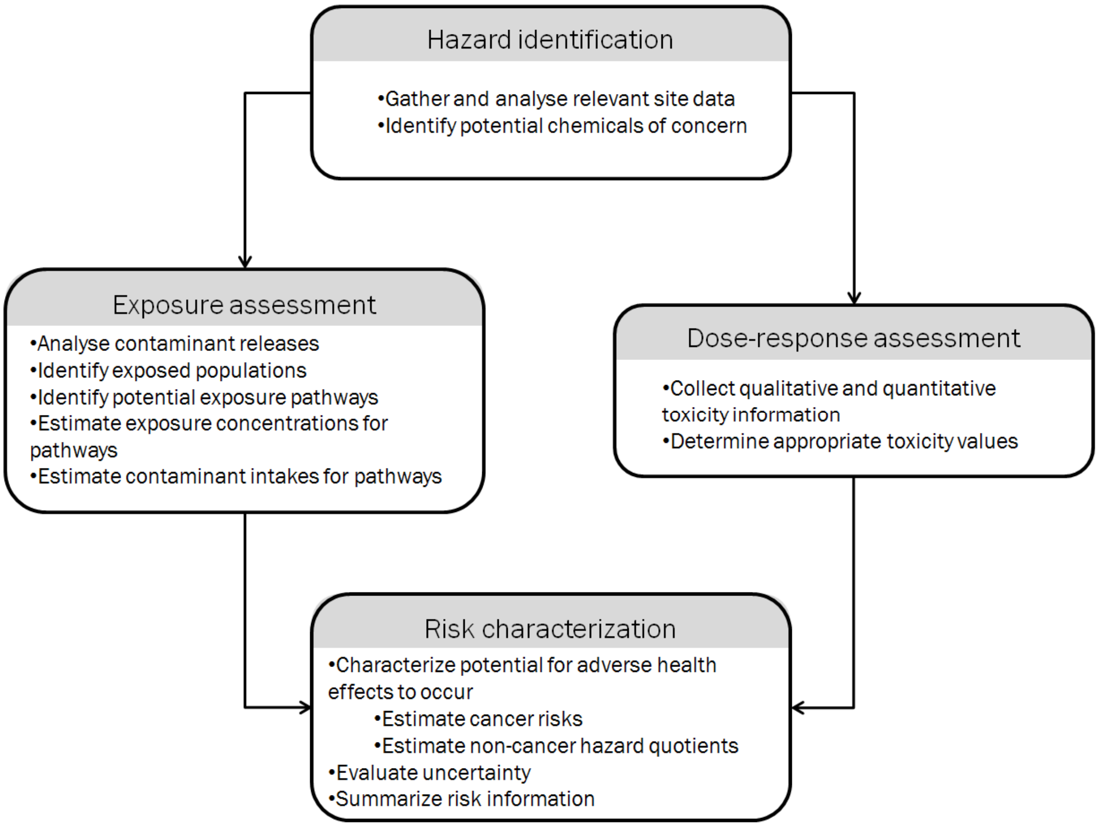

Figure 2.1. Framework for risk assessment. Adapted from USEPA (1989).

Data collection and evaluation involves gathering and analyzing the site data relevant to the human health evaluation and identifying the substances of concern.

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The exposure assessment estimates the magnitude of actual and/or potential receptor exposures to environmental contaminants, the frequency and duration of these exposures, and the pathways by which the risk group may be exposed (Asante-Duah, 1998).

The toxicity assessment considers the type of adverse health effects associated with chemical exposures, the relationship between magnitude of exposure and adverse effects, and related uncertainties. Typically, these data is obtained in lab studies, where toxicity information is gathered for specific chemicals.

Finally, the risk characterization combines the outputs from the exposure and toxicity assessments to characterize the risk. In this phase, the toxicity information is compared with the exposure levels to determine current and/or future risk levels.

Several health risk assessment methods are described in the literature (Franco et al., 2006; Legind and Trapp, 2009; Margni et al., 2004; Nadal et al., 2009b). Probabilistic risk assessment (PRA) is a general term for risk assessments that use probability models to represent the likelihood of different risk levels in a population (variability) or to characterize uncertainty in risk estimates (USEPA, 2001).

While point estimate risk assessment gives a central tendency of exposure or a maximum exposure estimate of risk, PRA determines the probability distribution of risks, reflecting the variability and/or uncertainty related to all input parameters. One of the most widely used methods for PRA is the Monte Carlo analysis (Schuhmacher et al., 2001).

Monte Carlo analysis is a statistic sampling technique that uses computer simulation to generate a great amount of input scenarios (samples), taking into account the information of the variability and uncertainty related to these input parameters. Each model run gets one of these input scenarios and calculates one output scenario, through the exposure and risk equations. The simulation gives the results in the form of a probability distribution function around a mean value and allows the carrying out of a detailed sensitivity analysis.

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For the calculation of human health risks, it is important to understand the concept of dose-response relationships. The relationship between the degree of exposure to a chemical (the dose) and the magnitude of a chemical-induced effects (the response) is described by a dose-response curve. A schematic representation of dose-response curves is given in Figure 2.2.

It may be noted in Figure 2.2 that for non-threshold chemicals (A) no minimum level is required to induce adverse toxicity effects. This is the case of most carcinogens, since these contaminants may represent damage even in extremely low doses.

In contrast, a low dose of threshold chemicals (B in Figure 2.2) causes no observable effects whereas a higher dose will result in some toxicity, and still higher doses cause even greater toxicity – up to the point of fatality. Non-carcinogens generally fall into this group. Non-cancer or systemic toxicity is generally treated as if there is an identifiable exposure threshold below which there are no observable adverse effects. The most important part of a dose-response curve with a threshold chemical is the dose at which significant effects first begin to occur. The highest dose which does not produce an observable adverse effect is called the 'no-observed-adverse-effect level' (NOAEL), and the lowest dose that produces an observable adverse effect is the 'lowest-observed-adverse-effect level' (LOAEL).

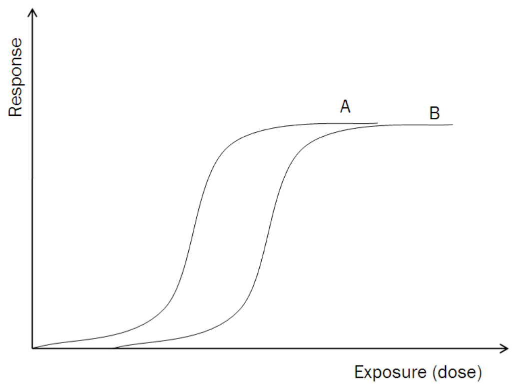

Figure 2.2. Schematic representation of exposure-response relationships: Illustration of dose-response relationship for non-threshold chemicals (A) and threshold chemicals (B).

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To be applied on risk calculation, a substance's dose-response relationship must be defined numerically by its toxicity value. The toxicity values defined by USEPA (1989) are the slope factor (for carcinogenic effects) and the reference dose (for non-carcinogenic effects). The slope factor (SF) is an "upper-bound estimate of the probability of a response per unit intake of a chemical over a lifetime". The chronic reference dose (RfD) is an "estimate of a daily exposure level for the human population, including sensitive subpopulations, that is likely to be without an appreciable risk or deleterious effect during a lifetime".

Given this context, human exposure to a contaminant is defined as (Equation 2.4):

$$CDI = \frac{C \cdot IR \cdot EF \cdot ED}{BW \cdot AT} \quad (2.4)$$

Where CDI is the chronic daily intake of the chemical (mg kg^{-1} day^{-1}), C is the concentration of the chemical in an exposure medium (mg L^{-1} or mg kg^{-1} dw), IR is the ingestion rate (L day $^{-1}$ for water, kg day $^{-1}$ for food, etc), EF is the exposure frequency (days $year^{-1}$), ED is the exposure duration (years), BW is the body weight (kg) and AT is the averaging time (days).

Cancer risk (CR) is calculated as (Equation 2.5):

$$CR = CDI \cdot CSF \quad (2.5)$$

Where CSF is the cancer slope factor (linear low-dose cancer potency factor) for the chemical (kg day mg^{-1}).

Non-carcinogenic risk or hazard quotient (HQ) is defined by Equation 2.6.

$$HQ = \frac{CDI}{RfD} \quad (2.6)$$

Where RfD is the reference dose for the chemical for assessing non-cancer health effects (mg kg^{-1} day^{-1}).

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2 Decision models

Decision and information support tools represent a broad and diverse category of computer-science based instruments (McIntosh et al., 2008). The application of these instruments requires prior knowledge and understanding of the problems, processes and activities leading to a decision. Decision tools provide means for structuring and exploring problems, generating qualitative and quantitative information to analyse and characterize these problems. The increasing interest on environmental management tools lies on the complexity and uncertainty related to natural systems.

Modelling sustainable development implies management of interdependencies between multiple and conflicting goals, a search for solutions that are equitable to current and future generations, and assessment of potential and chronic threats and protection from counterproductive disruption (Bui, 2000).

Kersten (2000) proposes a classification of decision support methodologies. Between those, multicriteria decision analysis (MCDA) and Bayesian networks (BN) were selected to be applied in this thesis. These methods are described in the following sections. MCDA is used to select and combine decision attributes, define objectives and goals, determine preferences and trade-offs among objectives and utility or value functions. They are also used to determine preference consistency and remove cognitive biases by the consistent use of preferences and tradeoffs. In turn, BNs are applied to identify and remove systematic inconsistencies and biases that arise from human cognitive limitations. They include aids to structure decision problems, estimate probability distributions, analyze risk and check for consistency of the decision maker's reasoning (Kersten, 2000).

2.1 Multicriteria Decision Analysis

Multicriteria Decision Analysis (MCDA) has been largely applied to support environmental decision making in the last few years (Aragonés-Beltrán et al., 2010, Browne et al., 2010, Crane et al., 2010, Fealy et al., 2010, Li et al., 2010, Paterson et al., 2010, Sorvari and Seppälä, 2010). According to Giove et al. (2009), MCDA is

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especially useful for environmental management problems, which require balancing scientific findings with multifaceted, value-laden input from many different stakeholders with different priorities and objectives.

The use of MCDA allows integrating different criteria (i.e., environmental, social, economic, etc.) that are measured in diverse scales and sometimes incommensurate. The selected criteria shall: (i) be able to discriminate among the alternatives and support the comparison of the performance of the alternatives, (ii) include all important aspects of the objectives, (iii) be sufficiently concise and non-redundant, and (iv) be measurable either in a quantitative or qualitative scale.

The MCDA classification is briefly described as follows (adapted from Vincke, 1992):

- MAUT/MAVT (Multi-Attribute Utility/Value Theory): The criterion values, normalized into a common numerical scale by means of a suitable transformation function (utility or value function), are aggregated by means of an operator, a function which satisfies a set of rationally axioms.

- Outranking: The method is based on an “outranking relationship”, stating that an alternative may have a degree of dominance over another one. Relationships are neither complete nor transitive. In this way, only a partial ordering is produced, implicitly admitting that comparable alternatives may exist.

The case-study here presented in this thesis has no comparable alternatives, as it is based on the selection of the most suitable areas to amend with sewage sludge. Therefore, a MAUT approach was selected.

The selection of the most suitable aggregator for the calculation is still a subjective issue. For the present case-study, a method called Logic Scoring of Preference (LSP) was found to be the most suitable to reach the thesis objective.

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2.1.1 Logic Scoring of Preference

The Logic Scoring of Preference (LSP) method is based on mathematical models that use generalized conjunction/disjunction (GCD) and other continuous preference logic functions (Dujmovic, 2007). LSP operators take into consideration the different levels in the hierarchy of criteria and the user weights and constraints over those criteria. Preference scales can be defined using multiple linear segments and linear interpolation. After defining elementary criteria for all performance variables, elementary preferences are generated. The next step is to aggregate elementary preferences and to compute the global preference that reflects the global ability of the evaluated system to satisfy all the requirements.

The aggregation of partial preferences is based on generalized conjunction/disjunction (GCD) logic that is used for modeling simultaneity and replaceability (Dujmovic and Larsen, 2007). Dujmovic and Nagashima (2006) described basic logic aggregation operators (aggregators). Each of these aggregators has specific logic properties, and the operators can be combined in other ways to create a wide spectrum of logic relationships to reflect the user needs.

- Simultaneity aggregator (partial conjunction or full conjunction): used when a simultaneous high satisfaction of all requirements in a group is needed.
- Replaceability aggregator (partial disjunction or full disjunction): used whenever the high satisfaction of any requirement can (partially or completely) replace the satisfaction of all other requirements in the group.
- Neutrality aggregator (arithmetic mean): located between replaceability and simultaneity, it combines a moderate need for simultaneous satisfaction of requirements with a moderate replaceability capability.

The simultaneity, neutrality, and replaceability are three fundamental related operators that are special cases of the GCD function (symbol $\Diamond$ in Equation 2.7).

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$$E_1 \lozenge \ldots \lozenge E_k = \left(\sum_{i=1}^{k} W_i E_i^r\right)^{1/r}, -\infty \ll r \ll +\infty \tag{2.7}$$

$$0 < W_i < 1, \qquad i = 1, \ldots, k, \quad \sum_{i=1}^{k} W_i = 1$$

Where E_i represents the preference of each criterion *i*, W_i represent the weight associated to each criterion *i* (used to express the relative importance of input preferences) and *r* is the exponent (used to adjust logic properties of this aggregation function).

The fundamental property of the partial conjunction is "*andness*", defined as a level of similarity between the partial conjunction and the full conjunction. The fundamental property of the partial disjunction is *"orness"*, defined as the level of similarity between the partial disjunction and the full disjunction. GCD is a combination of conjunctive and disjunctive properties. In the case of partial conjunction, conjunctive properties predominate, and in the case of partial disjunction, disjunctive properties are predominant. In the case of neutrality (arithmetic mean), the conjunctive and disjunctive properties are perfectly balanced.

2.2 Bayesian Networks

Bayesian networks (BNs) have been also successfully applied in environmental decision problems, and especially on land management and land use policy. Cain et al. (2003) investigated whether Bayesian networks could provide the generic framework to develop a DSS for agricultural system management. BN were perceived to have helped the planning process as they allowed policy makers to develop complex systems from a multi-disciplinary perspective. Ticehurst et al. (2011) compared BN with conventional statistical analysis to explore the usefulness of BNs for the analysis of social data sets in a case-study of natural resources management. The authors found that BNs results were more easily interpreted and communicated than traditional statistical outputs. Also, Bacon et al. (2002) performed a two stage model of land use change. BNs were applied to explore the relations between personal satisfaction and the costs for the landholders to change their land use. The authors

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argued that BNs is a powerful and robust method to bridge the gap between stakeholders and experts.

An important advantage of this method is the ability of explicitly dealing with uncertainty (van Kouwen et al., 2008). BNs are able to integrate knowledge from different disciplines, aggregating system complexity to the level that is appropriate, representing and communicating uncertainties. The development of the models through influence diagrams that describe causal relationships between input parameters and indicators is another important asset. The application of this tool improves the understanding of the environmental decision problem, leaving the decision makers to reach their own conclusions on the basis of that understanding.

The method is based on Bayes' rule of probability. In Bayesian inference, all quantities are treated as random variables, some of them, denoted *Y*, being fixed at known values (data, covariates), and the others, denoted θ, being unknown (parameters, latent variables, missing data). The method is based on using the Bayes theorem to calculate the posterior distribution, $P(\theta|Y)$, of unknown quantities, θ, given known quantities, *Y*, from the distribution, $P(Y|\theta)$, of known quantities, *Y*, given unknown quantities, θ, also known as the likelihood(Equation 2.8).

$$P(\theta|Y) = \frac{P(Y|\theta)P(\theta)}{P(Y)} \quad or\ P(\theta|Y) \propto P(Y|\theta)P(\theta) \qquad (2.8)$$

The Bayes theorem also involves $P(\theta)$, which is termed the prior distribution, and contains prior knowledge about parameter values, whereas $P(Y)$ is a normalization term. Consequently, Bayesian inference can also be seen as operating the transition from the prior to the posterior probability of θ, taking likelihood into account (Billoir et al. 2008). The probability theory underlying BNs provides a consistent calculus for uncertainty inference, and so a BN-based integrated model can be a useful tool to analyse the implications of uncertainties for future decisions (Lerner et al., in press). A complete description of the method application is given by Cain (2001).

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3 Integration on Geographic Information Systems

Geographic Information Systems (GIS) are computer-based systems for the capture, storage, retrieval, analysis and display of spatial data (Skidmore, 2002). Due to its analytical capacity and the potential of managing a great amount of data, GIS have been increasingly used in environmental decision making. Malczewski (2006) reports an important increase in the number of publications relating GIS and MCDA between the years 1990 and 2004 (Figure 2.3). The author reports that agriculture is one of the major application areas of spatial decision making. In recent studies, Ceballos-Silva and López-Blanco (2003) applied these tools to identify suitable areas for the production of maize and potato crops. In addition, Morari et al. (2004) combined GIS and MCDA to select criteria for best management practices in agricultural sites.

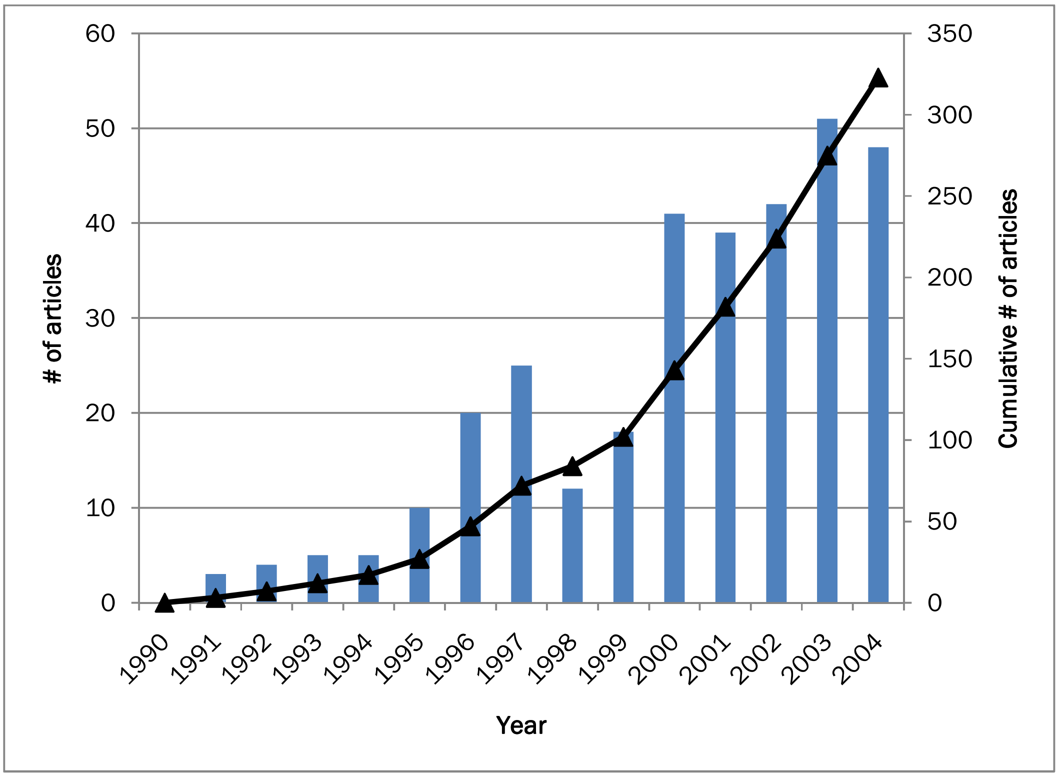

Figure 2.3. Total number of GIS-MCDA articles per year for the period 1990 – 2004. Based on Malczewski (2006)

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The number of articles that apply GIS coupled with BNs is much smaller (around 33 by searching "GIS" and "Bayes" as keywords in Scopus database). Most of these studies gather the data from GIS and apply these data to a constructed BN model.

GIS can be coupled with modelling, optimization or other methods (Sànchez-Marrè et al., 2008). These systems can be used iteratively with varying inputs and outputs representing spatial differences or changes. In this way, systems relationships could be explained. Sensitivity analyses may be also performed in a GIS basis.

4 Uncertainty incorporation in environmental models

Uncertainty is an inherent characteristic of environmental models, due to several reasons: (i) complexity of environmental systems, (ii) subjectivity related to environmental management problems, (iii) conflicting interests of a stakeholders, (iv) non-existence of the "perfect" alternative, etc. Although many sources of uncertainty are recognized, there is still a lack of information and agreement on their characteristics, magnitudes, and available means of dealing with them.

Ascough II et al. (2008) classified uncertainty in three typologies: knowledge uncertainty, variability uncertainty and linguistic uncertainty (Figure 2.4). Knowledge uncertainty is also known as epistemic or epistemological uncertainty. It refers to the limitation of knowledge, and may be reduced by additional research and empirical efforts. Variability uncertainty is related to the inherent variability of natural and human systems. Finally, linguistic uncertainty arises as a consequence of a vague, ambiguous and context dependant communication of the problem.

Even though uncertainty is inherent, it must not be rejected, as uncertainty representation gives a broader understanding of the problem. There are two main forms of uncertainty representation. The first is called the frequentist assumption (Ascough II et al., 2008): a true, fixed value for each parameter of interest exists, and the expected value of this parameter is obtained by random sampling, and the underlying parameter distribution is known. Monte Carlo method (section 2.2) constitutes an example of this approach.

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An alternative approach is the Bayesian inference (section 2.5). This method provides a mechanism to quantify uncertainty in parameter estimates, and to determine the probability that an explicit scientific hypothesis is true, given a set of data.

Sampling methods, such as Monte Carlo, assume that the randomisation of experiments is plausible, which sometimes is not true as ecological experiments are rarely repeated independently. In contrast, BNs consider that a prior distribution of the hypothesis exists. In fact, there is not a preferred approach to solve problems. Both approaches may be used for inference and decision making, and each one may involve some subjective or technical difficulties (Press, 2005).

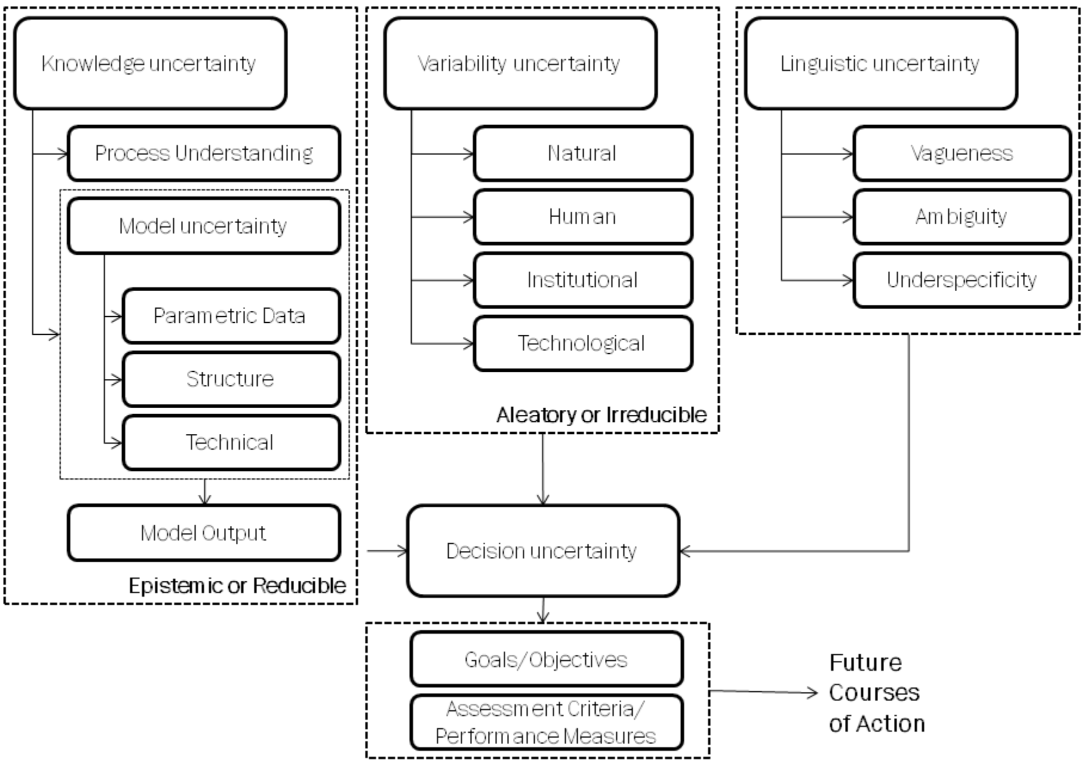

Figure 2.4. Uncertainty classification for environmental decision models. Based on Ascough II et al. (2008)

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5 Sensitivity analysis

Sensitivity analysis (SA) is the study of how the uncertainty in the output of a model can be apportioned to different sources of uncertainty in the model input (Saltelli, 2002). This analysis promotes a better understanding of how the models work, given the importance of each input. Due to this fact, this tool is mostly useful for model checking and calibration.

Campolongo et al. (2000) define three categories of SA:

- Factor screening, where the main task is the identification of influential factors in a system with many factors.
- Local SA, where the local (point) impact of the factors on the model is emphasized.
- Global SA focuses on apportioning the output uncertainty to the uncertainty in the input factors. Global SA typically takes a sampling approach, and the uncertainty range given in the input reflects our imperfect knowledge of the model factors.

A SA experiment is considered global when all the parameters are varied simultaneously and the sensitivity is measured over the entire range of each input parameter. This is the case that applies to this thesis.

Campolongo et al. (2000) describe sensitivity analysis methods and applications. Between those, regression analysis was selected to be applied in this thesis. Regression analysis is performed through the calculation of standardised regression coefficient (β), as follows (Equations 2.9 to 2.13).

$$\frac{y-\bar{y}}{\hat{s}} = \sum_j \beta \frac{x_j-\overline{x_J}}{\widehat{s_J}} \qquad (2.9)$$

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Where

$$\bar{y} = \sum_i \frac{y_i}{N} \tag{2.10}$$

$$\overline{x_j} = \sum_i \frac{x_{ij}}{N} \tag{2.11}$$

$$\hat{s} = \left(\sum_i \frac{(y_i - \bar{y})^2}{N-1}\right)^{1/2} \tag{2.12}$$

$$\hat{s}_j = \left(\sum_i \frac{(x_{ij} - \overline{x_j})^2}{N-1}\right)^{1/2} \tag{2.13}$$

N is the sample size (number of iterations in the Monte Carlo sampling method).

β ranges between zero and one. A value of β close to zero indicates that the output variable *y* is not correlated to input variable *x.* Moreover the sign of β also indicates the relationship between them, a positive β indicates that increments of the input variable *x*, are followed by an increase of the output variable *y*, and the opposite behaviour if the β is negative.

In this chapter, an overview of the methods applied to the thesis was presented. All these methods are suitable to be applied in different environmental management problems. The application of each of these methods to the case-study of SS amendment on agricultural soils is described in the following chapters.

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Part I

Environmental models

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Chapter 3

Development of a fugacity model to evaluate long-term contamination in soils amended with sewage sludge

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1 Introduction

Sewage sludge is the main residue of wastewater treatment plants (WWTP). Its application on agricultural fields is a managing practice of increasing use because of its benefits to soil and crops. This amending practice improves soil fertility, reduces the use of fertilizers, and is a relatively inexpensive solution. It also improves the soil structure by the addition of organic matter to the soil matrix and the nutrients recirculation through soil (Schowanek et al., 2004). In addition, the revalorization of a by-product (sewage sludge), which otherwise should be deposited in a landfill or incinerated, is achieved.

The Council Directive 86/278/CEE determines values for concentrations of heavy metals in soils to which sludge is applied, concentrations of heavy metals on sludge and the maximum annual quantities of such heavy metals which may be introduced into soil intended for agriculture. However, there are no legally enforceable restrictions related to organic compounds which may also be present in sludge. At the present day, limit concentrations for a series of persistent organic pollutants (POPs), such as polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons (PAHs), in sludge for use on land are only reported in the 3rd draft of the working document on sludge (CEC, 2000).

Sewage sludge can contain a wide range of organic compounds (Eljarrat et al., 2003; Katsoyiannis and Samara, 2005; Harrison et al, 2006) that may become transferred to sludge-amended agricultural soils. Persistent organic pollutants (POPs) are toxic, bioaccumulative and persistent chemicals, characterized by a subject to bioaccumulation potential and long-range transport capacity. These pollutants move through the different environmental compartments. Within these fate processes, POPs concentration dynamically changes over time and leads to accumulation on the food chain. As a final step, these contaminants may be transferred to the human diet.

Environmental models are tools for predicting pollutants distribution on the different media, and evaluate its risk for the population as well as the ecosystem, through

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exposure and risk assessment models. Among them, fugacity-based models have been largely used to predict the environmental distribution of POPs (Mackay and Fraser, 2000; Czub and McLachlan, 2004b; Wania et al., 2006). Fugacity is a convenient quantity to describe mathematically the rates at which chemicals diffuse, or are transported, between phases (Mackay, 2001), since the transfer rates are proportional to the fugacity difference between sources and destination compartments.

Fugacity models may be divided in four levels, according to their complexity (Mackay, 2001). Level I to III models assume that the system is at steady-state. Level I models also assume equilibrium in a closed system. Level II models include degrading reactions and advective loss, but all media are in equilibrium. On Level III models, the system is not in equilibrium. Level IV models are unsteady-state.

Amended soils have variable rates of emissions, since the applications are not continuous (on time) and are separated by large time intervals. In addition, the emission source is diffuse, impacting large spatial areas. In that case, Level IV models are the most appropriate fugacity models, as their equations are based on unsteady-state conditions. Moreover, these models are appropriate for chemicals that are very persistent and for which the time to steady state in the environment may be long (Arnot et al., 2008).

The main objective of this chapter was to develop and apply a multimedia model to predict the POPs accumulation on soil, after a long period of continuous sludge application. The behaviour of the contaminants and the influence of the soil matrix were particularly studied.

2 Materials and methods

2.1 Model description

Sewage sludge application on soil takes place at large time intervals, which are related to the cultivation time. The emission rate is not continuous in time, characterizing an unsteady-state condition. To calculate the rate of pollutants removal on soil, a dynamic fate model (Mackay Level IV) was employed (Hughes et al., 2005).

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The model input parameters were the following: sludge characteristics and application dose, soils characteristics (density, texture, organic matter content, transport parameters) and pollutant properties (solubility, k_{OW}, K_{OC}, temperature, vapour pressure, Henry and half-life in soil). Two layers of soil were considered (top and bottom layer). The soil matrix is composed by four compartments: air, water, organic matter and mineral matter.

The pollutant (*p)* mole balance differential equation for each soil compartment (*j)* is given by the Equation 3.1.

$$V_{ij} Z_{ijp} \frac{df_{ip}}{dt} = I_{jp} + \sum \left(D_{kp} f_{kp} \right) - \sum \left(D_{ip} f_{ip} \right) \tag{3.1}$$

Where V_{ij} is compartment volume (m^3 soil), Z_{ijp} is pollutant (*p)* fugacity capacity at the *same* compartment (mol m^{-3}·Pa^{-1}), f_{ip} is the pollutant (*p)* fugacity for the layer (*i)* (Pa). D_{kp} values represent the flow rates from layer *k* to layer *i* for pollutant *p* (mol- qp/Pa·h).

The fugacity of a contaminant *p* on each compartment *i* is proportional to the molar concentration (*M*) and inversely proportional to the bulk *Z* value (Mackay, 2001, Equation 3.2):

$$f_{ip} = \frac{M_p}{VZ_{Tip}} \tag{3.2}$$

The concentration (*C*) on each compartment *j* is described by Equation 3.3:

$$C_{ijp} = Z_{ijp} f_{ip} \tag{3.3}$$

The environmental compartments are linked by intercompartmental transfer processes. The processes considered in the model are depicted in Figure 3.1, and described as follows:

Loss processes in the top soil layer (1): volatilization (V) (D_{1Vp}) to the air above the top soil compartment, leaching (L) to the second layer (D_{1Lp}), sorbed phase transport (due

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to bioturbation B, D_{12Bp}) to the second layer, diffusion in pore air and pore water to the second layer (D_{12Ap} and D_{12Wp}) and biodegrading reactions (D_{1Rp}) (Equation 3.4).

$$V_1 Z_{1p} \frac{df_{1p}}{dt} = I_{1p} + \left(D_{21Bp} + D_{21Ap} + D_{21Wp}\right) f_{2p} +$$

$$+\left(D_{1Vp} + D_{1Lp} + D_{12Bp} + D_{12Ap} + D_{12Wp} + D_{1Rp}\right) f_{1p} \tag{3.4}$$

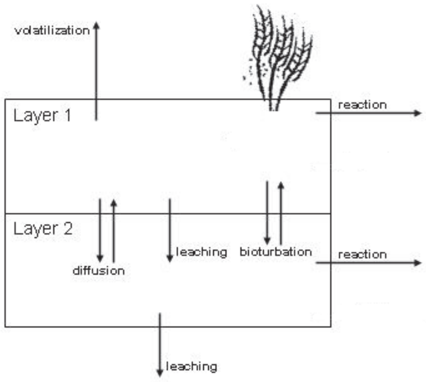

Figure 3.1. Transfer processes considered in the different environmental compartments. Adapted from Hughes et al. (2008).

Loss processes of the bottom layer (2), where the sorption (bioturbation, D_{21Bp}) and diffusion to the first layer through air and water compartments (D_{21Ap} and D_{21Wp}), leaching (D_{2Lp}), and degrading reactions (D_{2Rp}) (Equation 3.5).

$$V_2 Z_{2p} \frac{df_{2p}}{dt} = I_{2p} + \left(D_{1Lp} + D_{12Bp} + D_{12Ap} + D_{12Wp}\right) f_{1p} +$$

$$+\left(D_{2Lp} + D_{21Bp} + D_{21Ap} + D_{21Wp} + D_{2Rp}\right) f_{2p} \tag{3.5}$$

The global chemical loss (Equation 3.6) from the entire soil system is due to the chemical degradation in each layer (D_{1Rp} and D_{2Rp}), volatilization from the top layer (D_{1Vp}), and leaching from the lower layer (D_{2Lp}) as described in Webster and Mackay (2007).

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$$\frac{dM_p}{dt} = -\sum_{j\in 1\to out} D_{1jp} f_{1p} - \sum_{j\in 2\to out} D_{2jp} f_{2p} = -f_{1p}\left(D_{1Vp} + D_{1Rp}\right) - f_{2p}\left(D_{2Lp} + D_{2Rp}\right) \quad \forall p \qquad (3.6)$$

A fugacity approach was used in order to estimate the mechanisms of volatilization, diffusion, leaching, reaction, and bioturbation.

The model was completely developed within the MATLAB platform, taking into account data exchange into MS Excel. The mathematical model formulation requires solving a system of ordinary differential equations (ODEs). The time-depending variables are the pollutant species molar concentration in each compartment. As in total, there are two soil compartments and *J* compounds; the number of ODEs is *2J*. In this sense, the model is built to be able to accept any number of chemicals. The model is solved using MATLAB's solvers for initial value problems for ODEs. In this case, the algorithm that performs better, in computational time terms, was *ode23s*.

All model parameters regarding soil characteristics, chemical component physicochemical behavior and soil amendment features were stored in MS Excel for an ease of handling. These parameters were read into MATLAB using the *xlsread* MATLAB command. Model results such as temporal concentration profiles were sent back to MS Excel by using the *xlswrite* MATLAB command.

This MS Excel-MATLAB connectivity allows testing the model different parameters and performing parametric sensitivity analysis.

The model assumes that two soil layers may be differentiated. Each layer depth may be changed. Model calculation assumes that plant absorption is negligible and that transport values (*D*) are constant through the simulation time. These assumptions are related to POPs expected fate characteristics.

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2.2 Case-study

The model was applied to two types of Catalan (NE of Spain) soils, considering annual sewage sludge application. The first soil was characterized by having a sandy texture, a slightly acid pH, and a poor organic matter and nutrient content, while the second one had a sandy loam texture, a basic pH, and slightly higher organic matter content. The soil characteristics are summarized in Table 3.1.

Table 3.1. Soil parameters

Soil Parameter	Unit	Sandy	Sandy-loam
Organic matter content	%	1.10	2.30
Bulk density	kg m^{-3}	2425	2355
Molecular diffusivity in pore air	m^2h^{-1}	1.80E-02	1.80E-02
Molecular diffusivity in pore water	m^2h^{-1}	1.80E-06	1.80E-06
Diffusion distance on layer 1	m	5.00E-03	5.00E-03
Diffusion distance on layer 2	m	4.50E-02	4.50E-02
Air boundary layer thickness	m	4.75E-03	4.75E-03
Leaching rate	m h^{-1}	2.08E-04	2.08E-04
Bioturbation velocity	m h^{-1}	3.42E-07	3.42E-07
Layer 1 depth	m	1.00E-02	1.00E-02
Layer 2 depth	m	1.90E-01	1.90E-01

Several classes of chemicals may be found on sewage sludge (Eljarrat et al., 2003, Katsoyiannis and Samara 2005, Harrison et al. 2006). Among them, 10 polutants from 3 groups of chemicals (PCDD/Fs, PCBs and PAHs) were selected. These were chosen according to their hazard index (Nadal et al., 2008), which is based in turn on the persistent, bioaccumulation and toxicity potentials of the pollutants. The chemical parameters of the 10 POPs evaluated are detailed in Table 3.2. A 20-years simulation was performed to evaluate the long-term accumulation of the contaminants. This simulation considers that the soil is not contaminated (C_o=0). In addition, the field area (1 hectare) was supposed to receive 1 mmol of each contaminant on each annual amendment. It must be noted that these values do not correspond to a regular amendment, since the pollutant concentration in sludge and the initial amount in soil

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may substantially differ. However, they were selected to compare contaminants behaviour and also the differences between the two evaluated scenarios.

Table 3.2. Values of the 5 chemical parameters for the POPs evaluated (Mackay et al., 2000).

Chemical group	Chemical Name	MW[a] ($g{\cdot}mol^{-1}$)	Henry ($Pa\ m^{-3}{\cdot}mol^{-1}$)	$logK_{OC}$ [b]	$logK_{OW}$ [c]	Half-life in soil (d)
PCDD/Fs	2,3,7,8-TCDD	322	1.640	6.14	7.70	708
	OCDD	460	0.683	7.90	8.20	2329
	2,3,4,7,8-PeCDF	340	0.505	5.59	6.50	708
	OCDF	444	0.100	6.75	7.97	2292
PCBs	PCB 28	258	26.968	4.84	5.64	122[d]
	PCB 52	292	35.014	5.83	6.11	154[d]
	PCB 101	326	21.860	6.02	7.04	217[d]
PAHs	Acenaphthene	154	16.643	4.71	4.14	57
	Benzo(b)fluoranthene	252	1.223	6.24	6.39	485
	Fluorene	166	9.603	4.84	4.26	46

[a] MW: molecular weight, [b] K_{OC}: organic carbon partition coefficient, [c] k_{OW}: octanol-water partition coefficient, [d] source: Ayris and Harrad 1999

3 Results and Discussion

For all the studied pollutants, the removal profiles showed a tendency of reaching stability in terms of soil concentration through the simulation time (Figure 3.2). It could also be observed that as the contamination increased, the removal rates also increased, until reaching steady-state, when concentrations tend to remain constant with a constant sewage sludge application. This is explained by the fact that the higher concentrations the higher fugacity values, leading to a higher removal rate.

PAHs and PCBs showed a higher capacity of removal, for both scenarios evaluated. For these pollutants, the accumulation in soil was not observed. The only exception was

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benzo(b)fluoranthene, that presented a higher accumulation level than other PAHs, but much lower than PCDD/Fs.

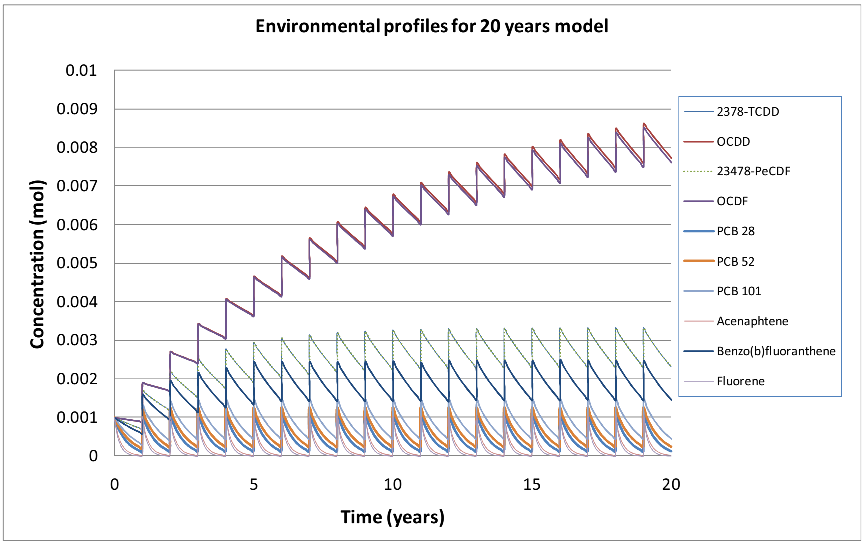

Figure 3.2. Temporal profile of the 10 POPs assessed for sandy soil.

In fact, PCDD/Fs were found to substantially accumulate in soils (Figure 3.2). As these contaminants are very lipophylic and tend to adsorb on soil organic matter, this trend was expected. The different degrees of accumulation are due to the different half-lives of these contaminants on soil. While 2,3,7,8-TCDD and 2,3,4,7,8-PeCDF have a half – life of two years approximately, OCDD and OCDF have a half-life of more than 6 years (Mackay et al., 2000). These different values conducted to a lower accumulation for 2,3,7,8-TCDD and 2,3,4,7,8-PeCDF, reaching a value of approximately 3 mmols after 4 years of annual application, and tending to become stable at this value. OCDD and OCDF need more time to become stable and reach stability on much higher values than the low- chlorinated PCDD/Fs.

The evaluated PAHs showed diverse behaviours through the simulation time (Figure 3.3). While acenaphthene and fluorene presented high removal rates, benzo(b)fluoranthene accumulated in the soil matrix, becoming stable on values between 1.5 and 2.5 mmol. This is probably due to the fact that benzo(b)fluoranthene has a more complex structure than the other evaluated PAHs, being more difficult to be

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degraded, and, consequently, presenting a higher half-life in soil. Similarly, high-substituted PCBs presented lower removal values (Figure 3.4).

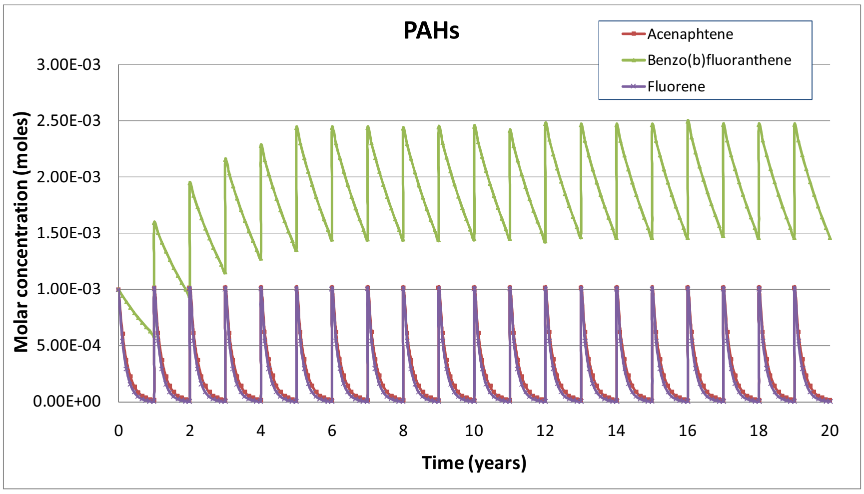

Figure 3.3. Temporal profile of PAH concentration in sandy soil in a 20-year model

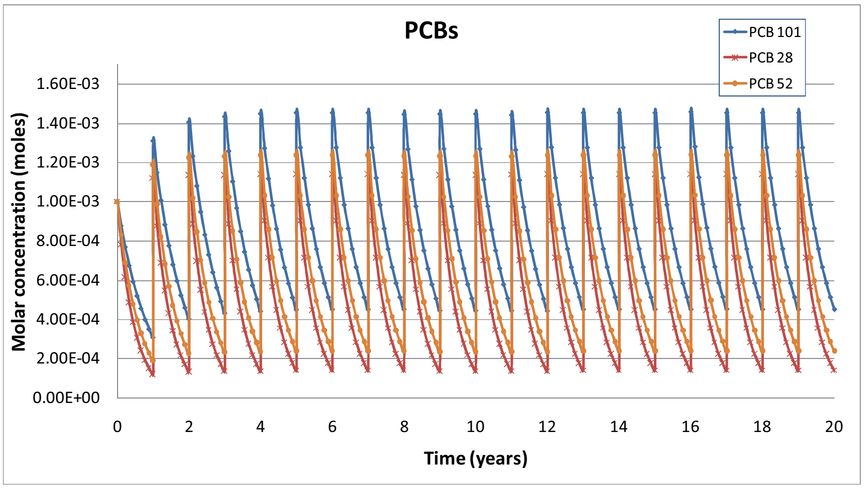

Figure 3.4. Temporal profile of PCB concentration in sandy soil in a 20-year model

No significant differences ($p>0.05$) were found between the two soils evaluated (Table 3.3). By the end of the 20 years, the sandy loam soil showed slightly lower concentrations for PAHs and PCB 101. The difference between the two scenarios for these contaminants, for a simulation period of 20 years, ranged between 1 and 3%, probably derived from differences in soils OM content.

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The contaminants with higher removal levels (PCB 28, PCB 52, acenaphthene and fluorene) were found to be more sensitive to soil parameters (Table 3.3). This may be explained by the fact that low half-life on soil leads to higher removal values, and the POP removal values are related to soil organic matter content.

In order to compare the chemical parameters and the removal profiles, two indexes were considered: a) accumulation rate - ratio between the final concentration and the annual contaminant charge (1 mmol)-; and b) removal rate - rate between the initial concentration (after amendment) and the final concentration (1 year after application). For the removal rate, the first year value was chosen since the removal rate is comparable only when contaminants have the same molar concentration (Table 3.4).

The statistic evaluation showed a high Pearson correlation ($r = -0.85$) between contaminants half-life on soil and the removal rate. An even higher correlation ($r > 0.99$) was observed between the accumulation potential and half-life on soil. For all other chemical parameters, only K_{OW} had a high correlation ($r = 0.93$) with the accumulation potential.

Table 3.3. Differences between the concentrations for the two evaluated soils types

	Final concentration (moles)		Difference [a]
Chemical Name	**Sandy soil**	**Sandy-loam soil**	**%**
2,3,7,8-TCDD	2.32E-03	2.33E-03	0.26%
OCDD	7.72E-03	7.72E-03	0.01%
2,3,4,7,8-PeCDF	2.31E-03	2.32E-03	0.45%
OCDF	7.62E-03	7.62E-03	0.04%
PCB 28	1.36E-04	1.40E-04	3.10%
PCB 52	2.34E-04	2.39E-04	1.88%
PCB 101	4.52E-04	4.46E-04	-1.24%
Acenaphthene	1.18E-05	1.15E-05	-2.91%
Benzo(b)fluoranthene	1.46E-03	1.46E-03	-0.45%
Fluorene	4.05E-06	3.96E-06	-2.51%

[a] *The negative values indicate that the removal rate is higher on sandy-loam soil than on sandy soil.*

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Table 3.4. Removal rate and accumulation potential of the assessed contaminants

Chemical Name	1st year removal rate	Accumulation rate
2,3,7,8-TCDD	30%	2.32
OCDD	10%	7.72
2,3,4,7,8-PeCDF	30%	2.31
OCDF	10%	7.62
PCB 28	88%	0.14
PCB 52	81%	0.23
PCB 101	69%	0.45
Acenaphthene	99%	0.01
Benzo(b)fluoranthene	41%	1.46
Fluorene	100%	0.00

4 Conclusions

The developed model allows predicting the behaviour of POPs in agricultural soils in the course of large periods of time. In fact, the model could be successfully applied to other groups of POPs, and even to different chemicals such as volatile compounds. This high flexibility is useful for supporting decisions related to sewage sludge management and its application on agricultural soils.

The model clearly showed the trends for each contaminant group, predicting a maximum concentration level that could be reached through continuous sewage sludge application. In consequence, the model results are useful to evaluate the environmental impact of amending soils, since the data exchanged with MS Excel platform can be easily connected to environmental risk models.

Regarding soil parameters, no significant difference was found between the two evaluated scenarios. However, an extended evaluation, which takes into account soils

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with higher organic content, must be carried out to check soil parameters participation on removal rates.

The statistical analysis showed that half-life in soil is the parameter having a key role. As this chemical property generally presents high standard deviation values for many chemicals, an uncertainty analysis needs to be performed for a better evaluation of the results. In addition, a sensitivity analysis is necessary to study the contribution of each parameter and the reliability of the results.

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Chapter 4

Sensitivity analysis applied to a fugacity model to evaluate long-term contamination in soils amended with sewage sludge

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1 Introduction

Persistent organic pollutants (POPs) are toxic, bioaccumulative and persistent chemicals with long-range transport capacity. These pollutants move through different environmental compartments (e.g., soil, water, air). Within these fate processes, POP concentration dynamically changes over time and leads to accumulation on the food chain. As a final step, these contaminants may be transferred to the human diet. There is evidence that sewage sludge can contain a wide range of organic compounds (Eljarrat et al., 2003; Katsoyiannis and Samara, 2005; Harrison et al., 2006), which can be transferred to sludge-amended agricultural soils. At present time, limit concentrations for a series of persistent organic pollutants (POPs), such as polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons (PAHs), in sludge for use on land have not been stablished, while they are only reported in the 3rd draft of the working document on sludge (CEC, 2000).

Environmental models are tools for predicting pollutants distribution on the different media compartments (e.g., soil organic matter, soil mineral matter, soil pore water, soil pore air). It is difficult to describe the fate of chemicals accurately, but the broad features of chemical fate can be understood and predicted when providing sufficient information on certain key chemical and environmental properties. These properties are partitioning properties, which control how the chemical is distributed between media, such as air and water, and reactive properties, which govern how fast the chemical reacts or degrades (Mackay et al., 2001).

Among environmental models, fugacity-based models have been largely used to predict the environmental distribution of POPs (Mackay and Fraser, 2000; Czub and McLachlan, 2004b; Wania et al., 2006). Fugacity is a convenient variable to describe mathematically the rates at which chemicals diffuse, or transport, between phases (Mackay, 2001), since the transfer rates are proportional to the fugacity difference between sources and destination compartments. Mackay proposes a classification of fugacity-based environmental models in different levels of increasing complexity: Level

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I to III models assume that the system is at steady-state, while Level IV considers dynamic modelling.

A very common destination of sludge sewage is to fertilize agricultural soils. In Spain, 65% of the sewage sludge produced in the year 2005 was disposed on agricultural fields. On the same period, in Catalonia, more than 160,000 tonnes of sludge were disposed on agricultural soils (MMA, 2008).

Amended soils have variable rates of emissions, since the applications are not time-continuous. However, the treatments are separated by large periods depending on the soil fertilization requirements. In addition, the emission source is diffuse. In this case, the most appropriate fugacity models are Level IV, as they are based on unsteady-state conditions and can manage these particularities. Moreover, these models are appropriate for environmentally persistent chemicals (Arnot et al., 2008). An essential point in environmental models is that chemicals partitioning and reactive properties vary enormously in magnitude from chemical to chemical, i.e. by a factor of a million or more, giving place to different chemical behaviours (Mackay et al., 2001).

As multimedia environmental models depend on several model input parameters to produce reliable results, a thorough analysis regarding the model sensitivity to the input parameter values deserves extra effort during their estimation. Sensitivity analysis (SA) is the study of how the variation in the output of a model can be apportioned, qualitatively or quantitatively, to different sources of variation and how the model output depends on input information (Campolongo et al., 2000). As a consequence, SA allows understanding the model insights.

The main objective of this chapter was to analyse the multimedia model described in Chapter 3. In addition, the effect of the uncertainty present in the model parameters on the prediction was studied.

2 Methodology

Various methods have been proposed to make uncertainty operational due to parameter uncertainty, such as the use of analytical uncertainty propagation methods,

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calculations based on intervals, applied fuzzy logic computation, and stochastic modelling describing parameters as uncertainty distributions (Huijbregts, 1998).

The use of analytical propagation methods suffers from complexity in algebra, leading to an increase of the model complexity. The model has a local perspective, as it provides information only on parameters sensitivity around the values which are slightly varied. Moreover, it is not accurate if uncertainties are large, the model is not smooth or important covariance terms are omitted. In this respect, Campolongo et al. (2000) pointed out that error propagation methods (i.e., derivative methods) provide only a local glimpse at model factors effect on model outputs. The use of sampling methods that explore the full space of possible model parameters values is recommended.

Any sampling methodology consists of three steps: (i) input variables scenario generation, (ii) model evaluation, and (iii) model output analysis. Several metrics can be calculated based on model results, such as standard statistics (mean, standard deviations, and confidence intervals) or regression analysis metrics. Sampling methodologies suffer from a severe problem, which rises from the lack of knowledge of the amount of scenarios required to generate statistically reliable output model distributions. In this sense, most methodologies adopt an iterative approach as follows:

i. To generate a certain number of scenarios which can be estimated based on previous mean estimations (Law and Kelton, 1999).
ii. To run the model for those scenarios.
iii. To calculate output model distribution parameters and SA metrics.
iv. To check that model output distributions compared to previous results are within specified tolerances. If not, return to (i).

The former methodology can be easily implemented for the case of Monte Carlo method where a new batch of scenarios can be easily generated without any

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requirement of being independent or dependent on prior generated scenarios, in contrast to Latin Hypercube Sampling (LHS) (Kurowicka and Cooke, 2006).

2.1 Standardized regression coefficients

Regression metrics are based on a linear correlation resulting from *N* input variables (*x*) and a given model output result (y_l). Standardization of input variables and output results is performed by subtracting the mean value and normalizing by the standard deviation (σ). This standardization procedure allows for magnitude comparison between variables, given that standardized variables have zero mean and a standard deviation of one. Standardized Regression Coefficients (SRC or β) are obtained using basic matrix inversion techniques (Chatterjee and Hadi, 1988). A value of any β close to zero indicates that the output variable is not linearly correlated to that input variable. The sign of β also indicates the relationship between input and output variables (Equation 4.1).

$$\frac{y_l-\bar{y}}{\sigma_y} = \beta\frac{x_l-\bar{x}}{\sigma_x} \qquad \forall l = 1:N \tag{4.1}$$

2.2 Partial correlation coefficients

Another commonly used metrics are Partial Correlation Coefficients (PCCs). These are calculated by performing several regressions including or not the variable under study (Equation 4.2). In this case, PCC calculates the contribution of each input variable on the behaviour of the output variables. This can be obtained by performing two separate regressions, one where all input variables are used (y_f^{all}) and another one where this specific input variable is ignored ($y_f^{\sim xh}$) (Helton and Davis, 2000).

$$PCC^2 = \frac{\Sigma\left(y_f-y_f^{xh}\right)^2-\Sigma\left(y_f-y_f^{all}\right)^2}{\Sigma\left(y_f-y_f^{xh}\right)^2} \tag{4.2}$$

PCCs have only positive values (range: 0-1). The values that are closer to one represent more sensitive variables, variables for which the regression without them makes the residuals larger.

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In the present study, a stochastic sampling approach was adopted. This means varying input data (model parameters) according to given probability distributions. The model was run for a given set of input values realizations (taken from the proposed probability distribution functions - *pdfs*), and its output results are stored. This procedure is repeated until the appropriate uncertainty ranges are obtained for the output variables.

In order to know which ones of the input variables mostly affect the model outputs, an iterative methodology was applied. The methodology, described in Helton and Davis (2000), consists on the following steps:

i. To calculate single variables SRCs for all the model input variables.

ii. To select the variable which has the highest absolute value of SRC, and for which the confidence interval does not contain zero.

iii. To subtract the explained variability by that variable from the output variable.

iv. To perform step (i) without the selected variables, until no more input variables are available, or when all remaining variables show SRCs which contain zero in their confidence interval.

The former procedure can help to rank model input variables impact on model output results. Note that PCCs can be also used with different ranking results.

3 Model application

3.1 Overview of the study system

The developed case represents the geographical region of Catalonia (NE Spain), and it is based on local landscape data and sewage sludge analysis. A single sewage sludge application was performed at the beginning of the time period. The considered soil was sandy, presenting low organic matter content (1.1%) and a slightly acid pH (6.8). After the application, sewage sludge was ploughed in a depth of 20 cm. Two soil layers are considered: the top layer has a depth of 1 cm while bottom layer has a depth of 19 cm. Soil parameters are presented in Table 4.1.

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As there are several classes of POPs on sewage sludge (Eljarrat et al., 2003; Katsoyiannis and Samara, 2005; Harrison et al., 2006), 10 different pollutants within three groups of chemicals (PCDD/Fs, PCBs and PAHs) were selected. They were chosen according to their hazard index (Nadal et al., 2008), which is based on the persistent, bioaccumulation and toxicity potentials of the pollutants. The chemical parameters of the 10 POPs evaluated are detailed in Table 4.2.

3.1 Sensitivity analysis

A Monte Carlo sampling was adopted. Scenarios were generated using the MATLAB random uniform and normal number generator functions (*unifrnd* and *normrnd*, respectively), the probability distribution function (*pdf*) for each parameter was selected based on previous information. In order to select the appropriate *pdf* for each model input parameter, the following criteria was used:

a. Literature survey of the parameter provides a single value: a uniform probability distribution is assigned considering ± 90% of the literature value.

b. Literature survey of the parameter provides five or less values: a uniform distribution function considering the minimum and the maximum value of those values is taken.

c. Literature survey of the parameter provides more than five values: a normal distribution function considering the mean and standard deviation of the value set is taken.

Given that normal distribution can cause negative parameter values if high standard deviations are defined, the absolute value of these is taken for the model run. Regarding uniform distributions the reason for such consideration is the complete lack of knowledge of other potential distributions, this *pdf* is considered to be the least informative and consequently the least biased.

In the case of soil parameters, all the selected distributions are uniform, with different boundaries selected according to previously mentioned criteria. Tables 4.2 and 4.3

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summarize the *pdfs* parameters for the model input variables considered to be uncertain, for soil and chemical parameters respectively. All remaining model variables were considered constant.

As previously mentioned, this sensitivity analysis is done considering one single amendment at the beginning of the first year, consequently and due to the ploughing action, both soil layers initially have the same pollutant concentration. No uncertainty is associated neither to the previously mentioned concentrations nor to the amendment amount. The model was run for a time span of 20 years. Model outputs studied were the soil chemical overall concentration and the removal rate.

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Table 4.1. Parameter distribuition for chemicals. Based on Mackay et al. (2000). (Continues)

Component	Henry constant (H_y) ($Pa\ m^{-3} mol^{-1}$)			Sorption partition coefficient (K_{OC}) (dimless)			Mineral matter-water partition coefficient (K_{MW}) ($L\ kg^{-1}$)		
	Distribution	Distribution parameters		Distribution	Distribution parameters		Distribution	Distribution parameters	
		Mean / Lower Bound	SD / Upper Bound		Mean / Lower Bound	SD / Upper Bound		Mean / Lower Bound	SD / Upper Bound
2,3,7,8-TCDD	Uniform[b]	2.8E+00	3.8E+00	Normal	6.1E+06	9.9E+06	Uniform[a]	8.0E-01	1.2E+00
2,3,4,7,8-PeCDF	Uniform[a]	4.0E-01	6.1E-01	Uniform[a]	3.2E+05	4.8E+05	Uniform[a]	8.0E-01	1.2E+00
OCDD	Uniform[a]	5.5E-01	8.2E-01	Normal	3.1E+07	4.3E+07	Uniform[a]	8.0E-01	1.2E+00
OCDF	Uniform[a]	8.0E-02	1.2E-01	Normal	7.9E+06	1.2E+07	Uniform[a]	8.0E-01	1.2E+00
PCB 28	Normal	2.7E+01	5.6E+00	Normal	6.9E+04	9.3E+04	Uniform[a]	8.0E-01	1.2E+00
PCB 52	Normal	3.5E+01	2.4E+01	Normal	6.8E+05	1.9E+06	Uniform[a]	8.0E-01	1.2E+00
Acenaphtene	Uniform[a]	1.3E+01	2.0E+01	Uniform[a]	4.1E+04	6.2E+04	Uniform[a]	8.0E-01	1.2E+00
Benzo(b)fluoranthene	Uniform[a]	9.8E-01	1.5E+00	Uniform[a]	1.4E+06	2.1E+06	Uniform[a]	8.0E-01	1.2E+00
Benzo(g,h,i)perylene	Uniform[a]	1.1E-01	1.7E-01	Uniform[a]	4.8E+06	7.3E+06	Uniform[a]	8.0E-01	1.2E+00
Fluorene	Uniform[a]	7.7E+00	1.2E+01	Uniform[a]	5.5E+04	8.3E+04	Uniform[a]	8.0E-01	1.2E+00

Distribution criteria:

[a] Literature survey of the parameter provides a single value: a uniform probability distribution is assigned considering ± 90% of the literature value.

[b] Literature survey of the parameter provides five or less than five values: a uniform distribution function considering the minimum and the maximum value of those five values is taken.

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Table 4.1. Parameter distribuition for chemicals. Based on Mackay et al. (2000) except for PCB's half lives on soil. (Continued)

Component	Octanol/Water Partition Coefficient (K_{OW}) (dimless)			Half life on soil (HLs_p) (days)		
	Distribution	Distribution parameters		Distribution	Distribution parameters	
		Mean / Lower Bound	SD / Upper Bound		Mean / Lower Bound	SD / Upper Bound
2,3,7,8-TCDD	Uniform[b]	4.7E+07	1.6E+08	Normal	4.2E+05	1.2E+05
2,3,4,7,8-PeCDF	Uniform[b]	3.0E+07	4.0E+07	Normal	4.2E+05	1.2E+05
OCDD	Uniform[b]	1.0E+12	2.6E+12	Normal	1.4E+06	2.4E+05
OCDF	Uniform[b]	9.5E+12	2.2E+13	Normal	1.3E+06	2.2E+05
PCB 28	Normal	4.4E+05	2.0E+05	Uniform[b]	6.7E+04	7.3E+04
PCB 52	Normal	1.3E+06	1.7E+06	Uniform[b]	8.3E+04	9.6E+04
Acenaphtene	Uniform[a]	1.1E+04	1.7E+04	Uniform[a]	4.7E+04	7.1E+04
Benzo(b)fluoranthene	Uniform[a]	2.0E+06	2.9E+06	Uniform[a]	2.8E+05	4.2E+05
Benzo(g,h,i)perylene	Uniform[a]	8.5E+06	1.3E+07	Uniform[a]	3.0E+05	4.5E+05
Fluorene	Uniform[a]	1.5E+04	2.2E+04	Uniform[a]	2.8E+04	4.1E+04

Distribution criteria:

[a] Literature survey of the parameter provides a single value: a uniform probability distribution is assigned considering ± 90% of the literature value.

[b] Literature survey of the parameter provides five or less than five values: a uniform distribution function considering the minimum and the maximum value of those five values is taken.

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The removal rate at different time-spans ($RemRate_{pt}$) for each pollutant was calculated as in Equation 4.3

$$Rem\ Rate_{pt} = \frac{M_{p0} - M_{pt}}{M_{p0}} \quad \% \qquad \forall p, t \tag{4.3}$$

where M_{pt} is the total mole amount of pollutant p at time t, while M_{p0} is the total mole amount of p at the initial time.

Table 4.2. Parameter distributions for soils

Variable description (Name)	Unit	Distri-bution	Distribution parameters		Source
			Lower Bound	Upper Bound	
Mass fraction of OM in soil OC (OC_{OM})	dimless	Uniform[a]	5.80E-02	1.10E+00	Webster and Mackay (2007)
Mass fraction of OC in soil solids in layer 1 (OC_{SS1})	dimless	Uniform[b]	9.56E-03	2.86E-02	Eljarrat et al. (2003)
Mass fraction of OC in soil solids in layer 2 (OC_{SS2})	dimless	Uniform[b]	9.56E-03	2.86E-02	Eljarrat et al. (2003)
Molecular diffusivity in pore air (B_A)	m^2/h	Uniform[a]	1.80E-03	3.42E-02	Webster and Mackay (2007)
Molecular diffusivity in pore water (B_W)	m^2/h	Uniform[a]	1.80E-07	3.42E-06	Webster and Mackay (2007)
Diffusion distance on top layer ($diff_1$)	m^2/h	Uniform[a]	5.00E-04	9.50E-03	Webster and Mackay (2007)
Diffusion distance on bottom layer ($diff_2$)	m^2/h	Uniform[a]	4.50E-03	8.55E-02	Webster and Mackay (2007)
Air boundary layer thickness (Y_B)	mm	Uniform[a]	4.75E-04	9.03E-03	Webster and Mackay (2007)
Leaching rate (L)	m/h	Uniform[a]	2.08E-05	3.96E-04	Webster and Mackay (2007)
Bioturbation velocity (V_B)	m/h	Uniform[a]	3.42E-08	6.50E-07	Eljarrat et al. (2003)

Distribution criteria:
[a] Literature survey of the parameter provides a single value: a uniform probability distribution is assigned considering ± 90% of the literature value.
[b] Literature survey of the parameter provides five or less than five values: a uniform distribution function considering the minimum and the maximum value of those five values is taken.

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4 Results and discussion

The number of scenarios run was varied and determined iteratively as proposed in section 2. Ten thousand scenarios were generated, and the results are shown in Figure 4.1. Regression based coefficient of multiple determination R^2 value, which provides a measure of the extent to which the regression model can match the observed data, was used to analyze the results.

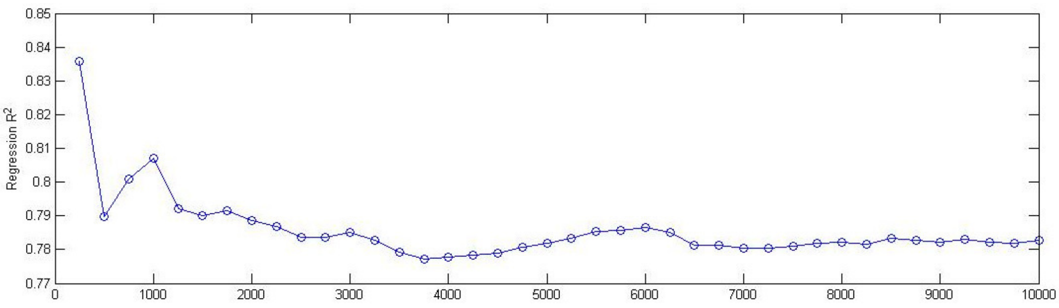

Figure 4.1. Regression's R^2 dependence on number of scenarios

The model output results were obtained by applying each of the sampled scenarios to the model described in Chapter 3. After approximately 3,000 scenarios the R^2 value remains around 0.78 (Figure 4.1), so this number of scenarios was selected.

Table 4.3 shows the average removal rates and average concentrations for each pollutant for different time spans. In all cases, the initial amount of each pollutant was fixed to be the same, and calculated after the amendment was done.

The calculation of SRCs and PCCs for the removal rate for different time spans was performed for all pollutants. The model results for soil and chemical parameters, in a time span of 5 years, are summarized in Tables 4.4 and 4.5, respectively.

Half life (HLs_p) presented the highest PCCs and SRCs values, being the most important factor. Other important variables were found to be mass fraction of organic carbon in soil organic matter (OC_{OM}), mass fraction of organic carbon in soil solids in layer 2 (OC_{SS2}), leaching rate (L) and octanol-carbon partition coefficient (K_{OC}) (in bold). The sign of SRCs indicate the correlation between the inpit parameters and the final

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concentrations. For positive SRCs higher values of those variables rendered higher final concentrations. This was the case of pollutant half lives (HLs_p), octanol-carbon partition coefficient (K_{OC}) and mass fraction of organic carbon in soil solids in layer 2 (OC_{SS2}). In contrast, for negative SRCs, higher values of these variables rendered lower final concentrations. This was the case of the mass fraction of organic carbon in soil organic matter (OC_{OM}) and leaching rate (L). These overall trends are the expected behaviour for such input parameter changes.

Table 4.3. Model estimation of mean pollutant removal rates (%) at different time intervals.

Time Interval considered	2,3,7,8-TCDD	2,3,4,7, 8-PeCDF	OCDD	OCDF	PCB 28	PCB 52	Acena-phtene	Benzo(b) fluoran-thene	Benzo (g,h,i) perylene	Fluor-ene
t=1 month	3.1	3.2	0.9	0.9	16.5	12.7	26.0	5.3	5.0	37.4
t=1 year	31.8	32.1	10.2	11.0	88.8	81.4	90.9	42.4	40.7	97.4
t=5 years	83.1	83.5	41.5	44.0	100	100	100	86.9	85.5	100
t=10 years	96.5	96.7	65.4	68.3	100	100	100	97.1	96.6	100
t=15 years	99.1	99.1	78.4	80.8	100	100	100	99.2	99.0	100
t=20 years	99.8	99.8	87.0	89.0	100	100	100	99.8	99.7	100

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Table 4.4. Standardized regression coefficients and partial correlation coefficients for soil parameters

Variable	2,3,7,8-TCDD		2,3,4,7,8-PeCDF		OCDD		OCDF		PCB 28		PCB 52		Acenaphtene		Benzo(b) fluoranthene		Benzo(g,h,i) perylene		Fluorene	
	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC
OC_{OM}	**-0.12**	**0.57**	**-0.11**	**0.60**	**-0.46**	**0.72**	**-0.44**	**0.70**	**-0.14**	**0.26**	**-0.16**	**0.51**	-0.02	0.03	-0.07	**0.35**	-0.07	**0.38**	-0.03	0.03
$OCSS_1$	0.00	0.03	0.01	0.04	0.02	0.04	0.01	0.03	0.03	0.05	0.02	0.08	0.01	0.02	0.00	0.02	0.00	0.03	0.00	0.00
$OCSS_2$	0.04	**0.25**	0.05	**0.31**	**0.16**	**0.34**	**0.16**	**0.34**	**0.13**	**0.24**	0.06	**0.23**	0.04	0.06	0.02	**0.13**	0.02	**0.14**	0.02	0.03
B_A	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.01	-0.07	0.14	-0.02	0.09	0.00	0.01	0.00	0.01	0.00	0.01	0.00	0.01
B_W	0.00	0.00	0.00	0.00	0.00	0.01	0.01	0.01	0.00	0.01	0.00	0.00	0.01	0.01	0.00	0.01	0.00	0.02	0.00	0.00
$diff_1$	0.00	0.01	0.00	0.03	0.00	0.00	0.00	0.00	0.06	**0.11**	0.03	**0.11**	0.01	0.01	0.00	0.02	0.00	0.00	0.00	0.00
$diff_2$	0.00	0.02	0.00	0.00	0.00	0.01	0.00	0.01	0.03	0.05	0.00	0.01	0.01	0.02	0.00	0.01	0.00	0.02	0.00	0.00
Y_B	0.00	0.01	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.01	0.01	0.00	0.02	0.00	0.01	-0.01	0.01
U_L	0.00	0.01	-0.02	0.13	-0.01	0.01	-0.01	0.02	**-0.13**	**0.23**	-0.01	0.06	-0.03	0.04	0.00	0.01	0.00	0.00	-0.03	0.03
V_B	0.00	0.01	0.00	0.00	0.00	0.01	0.00	0.00	-0.04	0.07	-0.01	0.02	-0.01	0.01	0.00	0.01	0.00	0.01	0.01	0.01

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Table 4.5. Standardized regression coefficients and partial correlation coefficients for chemical parameters

Variable	2,3,7,8-TCDD		2,3,4,7,8-PeCDF		OCDD		OCDF		PCB 28		PCB 52		Acenaphtene		Benzo(b) fluoranthene		Benzo(g,h,i) perylene		Fluorene	
	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC	SRC	PCC
H_y	0.00	0.00	0.00	0.00	0.00	0.01	0.01	0.01	-0.03	0.06	-0.03	0.11	-0.01	0.02	0.00	0.02	0.00	0.01	0.01	0.01
K_{OC}	0.01	0.04	0.03	0.21	0.01	0.03	0.02	0.04	**0.37**	**0.58**	0.07	**0.27**	0.05	0.08	0.00	0.03	0.00	0.00	0.05	0.06
K_{MW}	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.01	0.02	0.00	0.00	-0.01	0.01	0.00	0.02	0.00	0.00	-0.01	0.01
K_{OW}	0.00	0.01	0.00	0.00	0.00	0.00	0.01	0.02	-0.01	0.01	0.00	0.00	-0.01	0.01	0.00	0.00	0.00	0.01	0.01	0.01
HLs	**0.98**	**0.99**	**0.98**	**0.99**	**0.75**	**0.86**	**0.76**	**0.86**	**0.72**	**0.81**	**0.95**	**0.96**	**0.72**	**0.72**	**0.98**	**0.98**	**0.98**	**0.99**	**0.60**	**0.61**

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Concerning SRCs regressions, correlation R^2 coefficients were found to be above 0.70, and in some cases up to > 0.90 (Table 4.6). This is a quantitative proof that indicates that most model input variables act linearly and higher order interaction effects are not important. In this sense, it can be clearly stated that the pollutants behaviour depends mainly on their chemical properties and not on soil parameters, as linear relationships explain model output variability. Moreover the low values found for most input variables show that the model results greatly depend on a small group of parameters, rather than on the whole set of 15 variables. Afterwards, an iterative methodology has been applied (Table 4.6).

Table 4.6. Standardized regression R^2's values for different pollutants

R^2 values for SRCs	**2,3,7,8-TCDD**	**2,3,4,7,8-PeCDF**	**OCDD**	**OCDF**	**PCB 28**	**PCB 52**	**Acena-phtene**	**Benzo(b) fluoran-thene**	**Benzo (g,h,i)-perylene**	**Fluorene**
	0.973	0.977	0.801	0.803	0.730	0.930	0.946	0.967	0.971	0.848

Table 4.7 presents the percentage of total output variance explained by the input variables. It can be noted that most model output variability was explained by each pollutant half life (variable HLs_p. The remaining variability can be explained by mass fraction of organic carbon in soil organic matter (OC_{OM}) and chemical pollutant's octanol-carbon partition coefficient (K_{OC}), among others. The former variables are the ones that will be typically selected in a multivariate analysis, for which significant variables are those that individually explain at least more than (100%/(number of variables) = 100%/15) 6.7% of the output variable variance. The mass fraction of organic carbon in soil solids in layer 2 (OC_{SS2}) and leaching rate (L) seemed to be of minor importance, as they both explained less than 3% of the total variance.

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Table 4.7. Input variables percentage of total output variance explained from stepwise regressions.

Parameters	Variables	2,3,7,8-TCDD	2,3,4,7 8-PeCDF	OCDD	OCDF	PCB 28	PCB 52	Acenaphtene	Benzo(b)-fluoranthene	Benzo (g,h,i)-perylene	Fluorene
	OC_{OM}	1.30	1.30	**21.10**	**19.20**	2.00	2.50	0.40	0.50	0.50	0.30
	OC_{SS1}	0.00	0.00	0.00	0.00	0.10	0.00	0.00	0.00	0.00	0.00
	OC_{SS2}	0.20	0.20	2.60	2.60	1.60	0.40	0.10	0.10	0.10	0.10
	B_A	0.00	0.00	0.00	0.00	0.50	0.10	0.00	0.00	0.00	0.00
Soil	B_W	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	$diff_1$	0.00	0.00	0.00	0.00	0.30	0.10	0.00	0.00	0.00	0.00
	$diff_2$	0.00	0.00	0.00	0.00	0.10	0.00	0.00	0.00	0.00	0.00
	Y_B	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	L	0.00	0.00	0.00	0.00	1.60	0.00	0.00	0.00	0.00	0.00
	V_B	0.00	0.00	0.00	0.00	0.10	0.00	0.00	0.00	0.00	0.00
	H_y	0.00	0.00	0.00	0.00	0.10	0.10	0.00	0.00	0.00	0.00
	K_{OC}	0.00	0.10	0.00	0.00	**14.20**	0.60	0.10	0.00	0.00	0.10
Chemical	K_{MW}	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	K_{OW}	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	HL_s	**95.80**	**96.00**	**56.40**	**58.50**	**52.30**	**89.30**	**93.90**	**96.20**	**96.60**	**84.40**

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5 Conclusions

It has been shown that the use of SRCs and PCCs enables the study of how the input parameter behaviour affects model output. The current procedure enables to focus attention only on a few model input parameters for further studying, instead of the original set.

The sensitivity analysis pointed out that the most influential variable in modelling POP concentration in amended soils, using a Level IV model, was each pollutant half life (HLs_p). Moreover, other influential parameters were mass fraction of organic carbon in soil organic matter (OC_{OM}), mass fraction of organic carbon in soil solids in both layers (OC_{SS1}, OC_{SS2}) and leaching rate (L). These findings result after applying a stepwise regression methodology to the model with a single application amendment.

The former results indicate that any model enhancement should be aimed at performing better modelling of those parameters. As the chemical half life is depended on temperature, humidity, and solar radiation, these three factors could be introduced separately.

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Chapter 5

Integrated fate, exposure and risk model regarding sewage sludge application on agricultural soils

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1 Introduction

Sewage sludge (SS) management is being currently an important problem in wastewater engineering. Sewage sludge is the main residue of wastewater treatment plants (WWTP). It comprises the sludge originated in treatment plants of domestic wastewater that is occasionally mixed with industrial wastewater and/or run-off rain water (Metcalf and Eddy, 2003). As a solution, the recycling of sludge for agricultural purposes is an alternative to handle the increasing quantities of sludge produced in recent years.

With appropriate use, the application of sewage sludge to agricultural fields has benefits to soils and crops. This amending practice improves soil fertility (Fytili and Zabaniotou, 2008), reducing the use of fertilizers, while it is a relatively inexpensive solution. However, the presence of contaminants in sewage sludge may result in risks to humans and ecosystems (Fytili and Zabaniotou, 2008; Smith, 2009). The transport of pollutants between the environmental matrices may lead to soil contamination, as well as indirect emissions to air and water.

The European legislation defines levels of heavy metals in sewage sludge and agricultural soils intended to be amended (CEC, 1986). However, it is not concerned on any other kind of impacts to humans and ecosystems, such as the human exposure to organic contaminants through different routes.

Persistent organic pollutants (POPs) are toxic and persistent chemicals, characterized by being subject to bioaccumulation potential and long-range transport capacity. These contaminants are present at different concentrations in sewage sludge (Eljarrat et al., 2003; Harrison et al., 2006; Katsoyiannis and Samara, 2005), and are transferred to the soil matrix, as soils have a high capacity to act as reservoirs of organic pollutants (Vives et al., 2008).

Environmental models are tools for predicting contaminants movement and distribution between the different media, and to evaluate the risk for the human health as well as the ecosystems, through exposure and risk assessment models. Among

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them, fugacity-based models are largely used tools to predict environmental distribution of POPs (Czub and McLachlan, 2004b; Mackay and Fraser, 2000; Wania et al., 2006). Fugacity is a convenient quantity for describing mathematically the rates at which chemicals diffuse, or are transported between phases (Mackay, 2001).

In recent years, studies on transfer of pollutants to the food chain have been considerable (Armitage and Gobas, 2007; Collins et al., 2006; Czub and McLachlan, 2004b; Franco et al., 2006, Kelly et al., 2007; Margni et al., 2004; Wania et al., 2006). Among crop models, the most comprehensive are those which consider root uptake, particle deposition and diffusion (plant-air and air-plant), as well as losses by growth and metabolism (Legind and Trapp, 2009; McKone and Maddalena, 2007; Trapp and Matthies, 1995).

The objective of the present chapter was to assess the human health risks due to long-term sewage sludge application on agricultural soils. For that, four models were integrated: a multimedia model to assess POP fate in soils, a plant model, a food chain model, and an exposure and human health risks model. Uncertainty propagation and sensitivity analysis were performed to evaluate the importance of the input parameters.

2 Materials and methods

The integrated model was structured in four parts (Figure 5.1). The models are connected by their fluxes (inputs and outputs). Each model generates an output (concentration in the related matrix), which is used as an input of the next model. For example, soil model generates the soil concentrations in the defined time span. The calculated concentrations feed the plant, food chain and exposure and risk models, considering the same time span. While uncertainty propagation was performed for all the considered models, the sensitivity analysis was only performed for the last two steps, as the main focus of this study was on risk analysis and the soil model has been extensively evaluated in the previous chapter.

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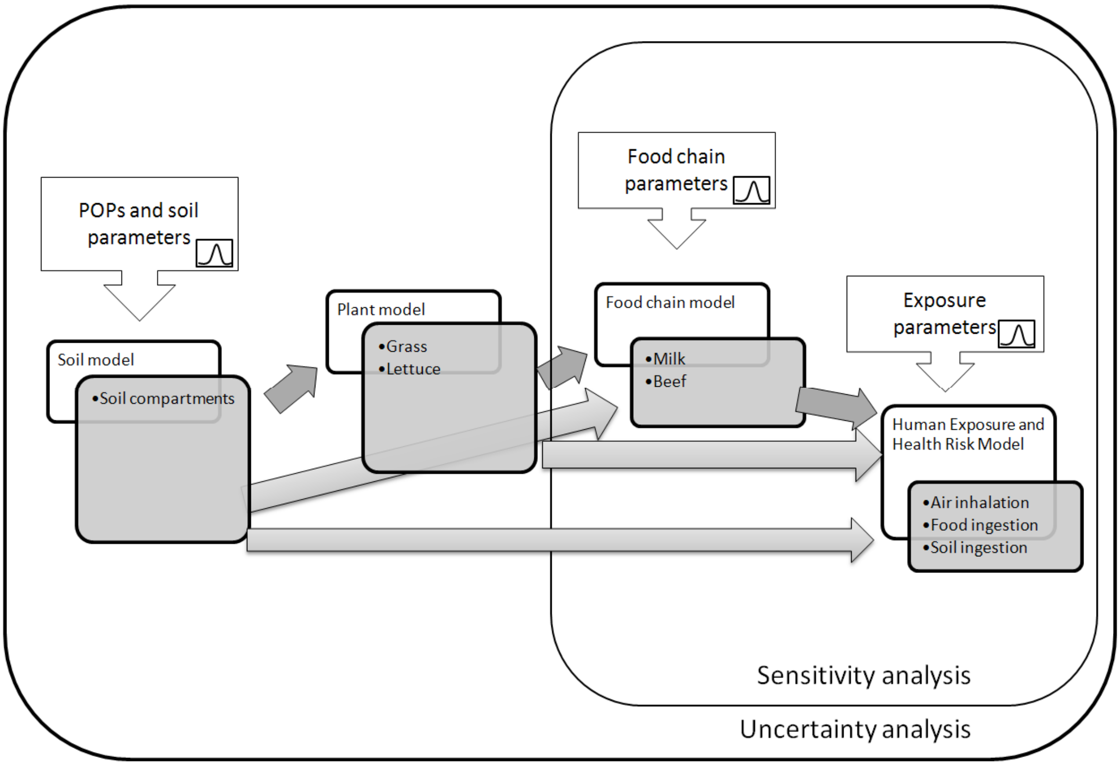

Figure 5.1. Schematic representation of the model development.

2.1 Soil model

Although details of the soil model have been previously given in chapter 3, some generalities are given in this section. In order to calculate the POP fate in the soil matrix, some peculiarities of this practice must be considered. Soil amendment occurs in determined periods of time, normally once or twice a year. Consequently, the emission is not continuous through time, characterizing an unsteady-state condition. Because of that, the rate of pollutants removal in soil was calculated through a dynamic fate model (Mackay level IV).

The model input parameters were: POP concentration in sludge, application dose, soils characteristics (density, texture, organic matter content, transport parameters), and pollutant properties (K_{OW}, K_{OC}, Henry constant and half-life on soil). The soil matrix is composed by four compartments: air, water, organic matter and mineral matter.

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The mass balance differential equation for a compartment *j* is decribed by Equation 5.1

$$V_{ij} Z_{ijp} \frac{df_{ip}}{dt} = I_{jp} + \sum \left(D_{kp} f_{kp} \right) - \sum \left(D_{ip} f_{ip} \right) \tag{5.1}$$

Where V_{ij} is the volume (m^3) of the compartment *j* in layer *i*, Z_{ijp} is the fugacity capacity (mol m^{-3} Pa^{-1}) in compartment *i*, f_{ip} is the fugacity (Pa) of the contaminant *p* on layer *i*, *t* is the time step (h), I_{jp} is the input rate (mol h^{-1}), D_{kp} represent the input transfers (mol $Pa^{-1}h^{-1}$) and $D_{ip}f_{ip}$ is the total output (mol h^{-1}).

The fugacity *f* of a contaminant *p* on each layer *i* is proportional to the contaminants molar concentration (M_p) and inversely proportional to the bulk value Z_{Tp} (Equation 5.2, according to Mackay, 2001).

$$f_{ip} = \frac{M_p}{V_i \times Z_{Tp}} \tag{5.2}$$

The molar concentration (M_{ijp}) of the contaminant *p* on each compartment *j* (mol m^{-3}) is described by Equation 5.3:

$$M_{ijp} = Z_{ijp} \times f_{ip} \tag{5.3}$$

The global chemical loss from the entire soil system is due to degradation in each layer, volatilization from the top layer, and leaching from the lower layer.

Within the soil model, POP physicochemical parameters and soil parameters were inserted as uncertain inputs. The result of this model was the distribution of the concentration of each contaminant in the soil matrix for the selected time span. This value fed the subsequent models (plant, food chain, and exposure and risk models).

2.2 Plant model

Two types of plants were considered: lettuce and pasture (grass). The contaminants fate from soil to plants was calculated considering three main routes: deposition, diffusion, and root transfer. Root transfer was calculated based on the concentrations of each contaminant in the soil matrix through time. For deposition and diffusion,

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equilibrium between soil and air was considered. The concentration in gas and particle was approximated by the chemicals octanol-air coefficient and organic matter (OM) fraction in the particle free to exchange chemical with the atmosphere, as described by Harner et al. (2000). The plant model is based on Legind and Trapp (2009). The molar concentration in leaf ($M_{L,p}$, on mol m^{-3}) is defined as (Equation 5.4):

$$M_{L,p} = \frac{Q_L \times TSCF \times C_{S,p} + A_L \times g \times C_{gas,p} + \dfrac{\upsilon_{dep} \times A_L \times C_{part,p}}{2}}{\dfrac{A_L \times g}{K_{LA}} + k \times V_L} \tag{5.4}$$

Where Q_L is the transpiration stream on leaves (m^3 d^{-1}), *TSCF* is the transpiration stream concentration factor (m^3 m^{-3}), $C_{S,p}$ is the concentration of the chemical p in soil (mol m^{-3}), A_L is the leaf area (m^2), g is the conductance for diffusive transfer between leaves and air (m d^{-1}), $C_{gas,p}$ is the concentration of the chemical p in the gas phase (mol m^{-3}), v_{dep} is the deposition velocity of particles on leaves (m d^{-1}), $C_{part,p}$ is the concentration of the chemical p in particle phase (mol m^{-3}), K_{LA} is the partition coefficient between leaves and air (unitless), k is the first order degradation rate (d^{-1}) and V_L is the area equivalent volume of above ground plant tissues (m^3). The concentration in leaves ($C_{L,p}$ in mg kg^{-1} dw) is defined by Equation 5.5.

$$C_{L,p} = \frac{10^3 \times M_{L,p} \times MW_p}{\rho(1-\upsilon_{W,L})} \tag{5.5}$$

Where MW_p is the molecular weight of the chemical p (g mol^{-1}), ρ is the leaf density (kg m^{-3}), $v_{W,L}$ is the volume fraction of water on leaves (unitless). A detailed description of the model is available from Legind and Trapp (2009). The model parameters were taken from Trapp and Matthies (1995).

For the plant model, soil concentration was considered as the only uncertain input. As a result, the concentrations in grass and lettuce were also represented as a distribution. The concentration distribution in grass fed the food chain model, while the concentration in lettuce fed the exposure and risk model.

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2.3 Food chain model

The concentration of POPs in cattle meat and milk, considering that pasture fields were amended with sewage sludge, was estimated by applying a food chain model. The parameters for calculating the levels were considered to be different for dairy cattle (milk) and beef cattle (beef). The concentrations in beef and milk were calculated based on the models described by Kumar et al. (2009), which is adapted from USEPA (1998). The input parameters of the food chain model are shown in Table 5.1. These parameters were taken from the literature, with the exception of the fraction of food intake by the animal from the polluted area (FI_{cattle}), for which USEPA (1998) indicates the use of a default value of 1. However, it was assumed that the amendment could be performed in a smaller area, as the portions of grain, silage and other feeds are not defined. Therefore, we considered that pasture (grass) represents more than a half of the cattle's diet. In addition, for the calculation of the biotransfer factor (B_a) in beef and milk, it was taken into account that log k_{OW} is truncated in a value of 6.9, as indicated by the USEPA (1998).

Table 5.1. Input parameters for the food chain model.

Parameters	Symbol	Units	Value *	Observation
Quantity of plant eaten by the animal	Qp_{milk}	kg day^{-1}	Uniform (2.6, 11)	Derived from data of seven types of grains, two types of forage and two types of silage, for beef and dairy cattle (USEPA 1997)
	Qp_{beef}	kg day^{-1}	Uniform (0.47, 8.8)	
Soil consumption rate (animal)	Qs_{milk}	kg day^{-1}	Uniform (0.1367, 2.64)	(1 – 18% of dry matter intake) USEPA (1997)
	Qs_{beef}	kg day^{-1}	Uniform (0.13, 1.17)	(1 – 18% of dry matter intake) USEPA (1997)
Fraction of ingested food from the amended area (cattle)	FI_{cattle}	unitless	Uniform (0.50, 1.00)	The consumption of food from amended area was assumed to be 50 – 100%

* The numbers in parenthesis indicate the minimum and maximum values of the distribution.

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2.4 Human exposure and health risk model

In order to evaluate the risks of amending soils with sewage sludge, 2 scenarios were defined. The first scenario considered farmers (occupational) exposure through diet, soil ingestion and particle inhalation. The second scenario contemplated exclusively non-occupational exposure through food consumption. Calculations were performed for adults. For that, only the ingestion of food from areas amended with sewage sludge was considered. The exposure data selected for the evaluation are shown in Table 5.2.

The exposure duration was defined as the same as the simulation time. Different values regarding the fraction of ingested food produced in the contaminated area were chosen depending on each scenario. It was considered that the percentage of local products in the diet of occupationally exposed populations ranged between 40% and 80%, while non-occupationally exposed population presented a lower proportion of food ingested from the area (between 10% and 30%). The exposure and risk model was based on Kumar et al. (2009) and USEPA (1998, 2009a).

2.5 Uncertainty and sensitivity analysis

For all the models, the variability of the parameters was taken into account. A 30-years simulation was executed, considering accumulation in both soil and the food chain. The uncertainty analysis was performed through the Monte Carlo method, with 5,000 iterations. The number of iterations was defined according to the coefficients of correlation (R^2). Finally, the sensitivity analysis was performed through the calculation of the standardized regression coefficients, β (Campolongo et al., 2000) for each uncertain input. The β values are effective for linear or quasi-linear models ($R^2 \geq 0.7$) and for prioritization purposes (Saltelli et al., 2008). β values were calculated with the software Analytica (Lumina, 2008). The use of this software allowed the integration of models and the analysis of the uncertainty. Also, it made easier the calculation of β values through the use of a toolbox called “Sensitivity Analysis Library”.

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Table 5.2. Input parameters for the human exposure model.

Parameters	Symbol	Units	Value	Reference
Soil ingestion rate	CR_s	mg day^{-1}	Triangular (0.1, 25, 50)[a]	Lagoy (1987)
Consumption rate of vegetables	CR_{veg}	g day^{-1}	Lognormal (99 ± 80)[b]	Arija et al. (1996)
Consumption rate of milk	CR_{milk}	g day^{-1}	Lognormal (226 ± 177)	Arija et al. (1996)
Consumption rate of beef	CR_{beef}	g day^{-1}	Lognormal (180 ± 84)	Arija et al. (1996)
Gastrointestinal absorption factor	AFI_g	unitless	Triangular (0.4, 0.6, 1.0)	Nessel et al. (1991)
Body weight	BW	kg	Lognormal (67.52 ± 12.22)	Arija et al. (1996)
Exposure frequency	EF	day yr^{-1}	Triangular (180, 345, 365)	Smith (1994)
Exposure time	ET	h day^{-1}	8	USEPA (2009a)
Exposure duration	ED	years	30	Unpublished data
Average lifetime	AT	years	Lognormal (75 ± 5)	Frey (1993)
Fraction of food produced in the amended area (occupational)	FI_{human}	unitless	Uniform (0.4, 0.8)[c]	Unpublished data
Fraction of food produced in the amended area (non-occupational)	FI_{human}	unitless	Uniform (0.1, 0.3)	Unpublished data

Note: The number in the parenthesis indicate
[a] The lower, mode and upper limit of a triangular distribution
[b] The mean and standard deviation of the lognormal distribution
[c] The minimum and maximum values of the uniform distribution

2.6 Case-study

The above described models were applied to a case-study: a Catalan (NE of Spain) sandy soil (slightly acid soil) with an organic matter content of 1.1% receiving 30 tons of sewage sludge (dw) per hectare, once a year. The selection of only one type of soil was based on a previous study (Passuello et al., 2009), where no significant differences were found between the removal rates for different types of soils.

The model was applied to 2 types of crops: lettuce and cattle fields. The food chain model considered two varieties of cattle (beef and dairy) that are exposed through ingestion of crops and soil.

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Four organic pollutants were selected: 2,3,7,8-TCDD, PCB 180, and 2 polycyclic aromatic hydrocarbons (PAHs): benzo(a)pyrene and dibenzo(a,h)anthracene. These contaminants were chosen according to their hazard index (Nadal et al., 2008) as well as the results of the previous chapters. The evaluated POPs were expected to present the highest human health risks of each chemical group.

The concentration of these contaminants in sewage sludge was taken from internal reports of the current research project. The results of this characterization were statistically treated and the mean concentration values were applied (Table 5.3).

Table 5.3. POP concentration in sewage sludge samples.

Chemical	2,3,7,8TCDD	Benzo(a)pyrene	Dibenzo(a,h)anthracene	PCB 180
Mean level (mg kg^{-1} dw)	7.51E-07	8.5E-02	2.0E-02	6.6E-02

As the objective of the present study was to determine the human health risks related to soil amendment, the initial soil concentrations were adjusted to zero and external sources were not taken into account, simplifying the model and reducing the computational effort of the calculation. The concentration in soil was calculated on a daily basis. The mean soil concentration for each year was used to estimate grass concentrations and soil ingestion. For lettuce concentration, it was considered that crops were harvested 70 days after sludge amendment. Non-cancer risk was calculated by comparing the exposure with the oral reference dose (RfD_o), while the cancer risk was established by multiplying the exposure and the oral slope factor (SF_o). Regarding inhalation, the cancer risk was derived by multiplying the exposure and the Inhalation Unit Risk (IUR), while non-carcinogenic risk was calculated by comparing the exposure through inhalation and the inhalation reference concentration (RfC_i). Risk parameters are summarized in Table 5.4.

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Table 5.4. Parameters for assessing cancer and non-cancer risks (USEPA, 2008, 2009b).

	Cancer risk		Non-cancer risk	
	SF_o	**IUR**	**RfD_o**	**RfC_i**
2,3,7,8TCDD	1.30E+05	38	1.00E-09	4E-08
Benzo(a)pyrene	7.3	1.1E-3	-	-
Dibenz(a,h)anthracene	7.3	1.2E-3	-	-
PCB 180	1.3	3.8E-04	-	-

Slope Factor Oral (SF_o) in kg day mg^{-1}, Oral Reference Dose (RfD_o) in mg kg^{-1} day^{-1}, Inhalation Toxicity Value (IUR) in m^3 μg^{-1}, Inhalation Reference Concentration (RfC_i) in mg m^{-3}.

3 Results and discussion

3.1 Soil model

The trends of the soil concentration of the selected contaminants for a period of 30 years are shown in Figure 5.2. The evaluated PAHs and PCB showed a nearly constant concentration through time and reached steady-state within the first years of sludge application. In addition, the uncertainty of the results for these pollutants did not show important changes through time. In contrast, 2,3,7,8-TCDD tended to accumulate in soils, because the time PCDD/Fs need to reach steady-state is higher (Passuello et al., 2009). By the end of the 30 years evaluation period, a tendency of reaching a stationary value was observed for all the contaminants. In comparison with data of the same region (Nadal et al., 2009a), PAH levels were approximately 10 times lower than those found for unpolluted soils, while the concentrations of chlorinated compounds (PCB 180 and 2,3,7,8-TCDD) were similar to those found by the same authors (Nadal et al., 2009a). Nevertheless, it should be noted that this comparison is only illustrative, as the present study did not consider the contamination from external sources.

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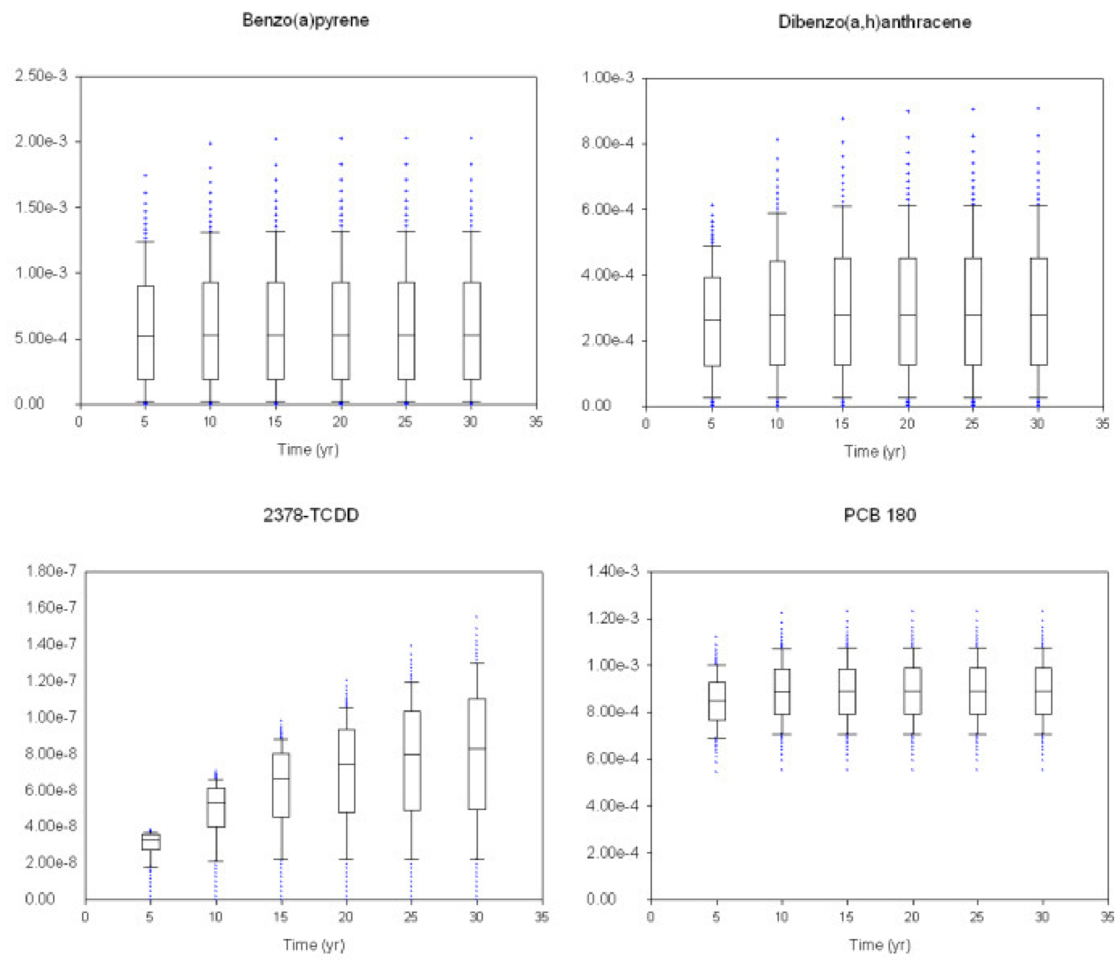

Figure 5.2. POP concentration in soil (mg kg^{-1} dw) along the simulation time. Upper and lower box boundaries indicate the 75th and percentile the 25th, respectively. The line within the box marks the median. Whiskers (error bars) above and below the box indicate the 90th and 10th percentiles, in this order. Points above and below the whiskers are outliers.

3.2 **Plant model**

Table 5.5 presents estimated POP levels in grass and lettuce. The concentrations in both types of plants were similar. As all the evaluated POPs have a high soil-air partition coefficient (K_{SA}), they tend to volatilize in tiny amounts. Among those, PCB 180 has the highest K_{SA}, tending to a lower volatilization and becoming attached to

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particles. In fact, for this contaminant, 72% of the inputs in plants came from particle deposition. In turn, 2,3,7,8-TCDD has the lowest K_{SA} among the assessed contaminants. Therefore, this chemical presented a higher volatilization level and a lower tendency to become attached to air particles. Diffusion plant-air for this contaminant represented 90% of the input fluxes. For the evaluated contaminants, root transfer was found to be the least contributing process (less than 1%).

The plant concentration profiles showed the same tendency than that observed in soil (Figure 5.2). This fact was expected as in the selected plant model (Equations 5.4 and 5.5) the concentration in plant ($C_{L,p}$) is related to the concentration in soil ($C_{S,p}$) and air ($C_{part,p}$ and $C_{gas,p}$). In addition, both $C_{part,p}$ and $C_{gas,p}$ were estimated according to $C_{S,p}$.

3.3 Food chain model

Table 5.5 shows the calculated values for concentration in meat and milk. Soil ingestion was found to be the most important route in meat and milk contamination, representing more than 99% of intake for cattle. Consequently, the uncertainty values were mainly related to uncertain concentrations in soil.

3.4 Exposure and risk model

The human exposure to 4 POPs through different pathways is presented in Table 5.6, while the associated cancer and non-cancer risks are shown in Table 5.7.

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Table 5.5. POP concentration in grass, lettuce, meat and milk (mg kg^{-1}), for a simulation time of 5 and 30 years.

			2,3,7,8-TCDD		Benzo(a)pyrene		Dibenzo(a,h)anthracene		PCB 180	
		Time	5 yr	30 yr	5 yr	30 yr	5 yr	30 yr	5 yr	30 yr
Grass		Mean	2.48E-10	6.69E-10	7.00E-07	7.43E-07	2.98E-07	3.57E-07	4.68E-07	4.92E-07
		SD	6.67E-11	3.00E-10	3.94E-07	4.25E-07	1.54E-07	1.95E-07	6.30E-08	7.44E-08
	Percentiles	50th	2.48E-10	6.63E-10	6.72E-07	7.10E-07	2.91E-07	3.44E-07	4.68E-07	4.92E-07
		90th	3.33E-10	1.06E-09	1.23E-06	1.32E-06	5.02E-07	6.18E-07	5.49E-07	5.88E-07
Lettuce		Mean	1.76E-10	7.00E-10	1.01E-06	1.22E-06	3.48E-07	5.08E-07	6.26E-07	8.11E-07
		SD	2.94E-11	2.91E-10	3.78E-07	5.39E-07	1.15E-07	2.32E-07	3.55E-08	7.70E-08
	Percentiles	50th	1.76E-10	6.97E-10	1.01E-06	1.21E-06	3.48E-07	5.03E-07	6.26E-07	8.11E-07
		90th	2.13E-10	1.07E-09	1.49E-06	1.92E-06	4.95E-07	8.10E-07	6.72E-07	9.09E-07
Meat		Mean	4.02E-09	1.08E-08	8.42E-05	8.68E-05	3.11E-05	3.51E-05	1.10E-04	1.17E-04
		SD	2.19E-09	7.20E-09	6.09E-05	6.28E-05	2.39E-05	2.63E-05	5.48E-05	5.77E-05
	Percentiles	50th	3.43E-09	8.77E-09	6.86E-05	7.32E-05	2.51E-05	2.56E-05	1.06E-04	1.13E-04
		90th	7.43E-09	2.16E-08	1.63E-04	1.75E-04	6.36E-05	7.96E-05	1.87E-04	1.92E-04
Milk		Mean	2.54E-09	7.05E-09	5.42E-05	5.63E-05	2.03E-05	2.48E-05	7.07E-05	7.43E-05
		SD	1.44E-09	5.05E-09	4.53E-05	4.62E-05	1.66E-05	1.97E-05	3.89E-05	4.05E-05
	Percentiles	50th	2.28E-09	5.27E-09	4.43E-05	4.82E-05	1.61E-05	2.09E-05	6.73E-05	6.99E-05
		90th	4.61E-09	1.38E-08	1.20E-04	1.25E-04	4.36E-05	5.28E-05	1.21E-04	1.23E-04

SD: Standard deviation.

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Table 5.6. Environmental exposure to the evaluated POPs (mg kg^{-1} day^{-1}) through different routes for the 2 assessed scenarios. Simulation time: 30 years.

		Occupational				Non-occupational			
				Percentiles				Percentiles	
		Mean	SD	50th	90th	Mean	SD	50th	90th
2,3,7,8-TCDD	Air inhalation	6.67E-12	2.72E-12	6.82E-12	1.07E-11				
	Soil ingestion	1.65E-14	1.12E-14	1.32E-14	3.41E-14				
	Food ingestion	1.72E-11	1.68E-11	1.21E-11	3.74E-11	5.82E-12	5.23E-12	3.57E-12	1.22E-11
Benzo(a)pyrene	Air inhalation	5.39E-09	2.05E-09	5.48E-09	7.80E-09				
	Soil ingestion	1.46E-10	1.18E-10	1.3E-10	2.92E-10				
	Food ingestion	1.43E-07	1.39E-07	1E-07	3.33E-07	4.59E-08	4.31E-08	3.3E-08	9.58E-08
Dibenzo(a,h)anthracene	Air inhalation	2.33E-09	9.66E-10	2.27E-09	3.51E-09				
	Soil ingestion	6.8E-11	5.38E-11	5.27E-11	1.33E-10				
	Food ingestion	5.94E-08	5.85E-08	3.58E-08	1.45E-07	1.97E-08	1.71E-08	1.5E-08	4.49E-08
PCB 180	Air inhalation	3.24E-09	6.39E-10	3.23E-09	4.13E-09				
	Soil ingestion	1.85E-10	1.12E-10	1.71E-10	3.07E-10				
	Food ingestion	1.87E-07	1.45E-07	1.41E-07	3.71E-07	6.12E-08	4.16E-08	5.07E-08	1.24E-07

SD: Standard deviation.

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Table 5.7. Carcinogenic and non-carcinogenic risks due to environmental exposure to the evaluated POPs. Simulation time: 30 years.

		Occupational				Non-occupational			
				Percentiles				Percentiles	
		Mean	SD	50th	90th	Mean	SD	50th	90th
Oral cancer risk	2,3,7,8-TCDD	6.86E-07	4.98E-07	5.57E-07	1.31E-06	2.26E-07	1.46E-07	1.8E-07	4.14E-07
	Benzo(a)pyrene	4.23E-07	3.34E-07	3.36E-07	8.43E-07	1.41E-07	1.03E-07	1.14E-07	2.91E-07
	Dibenzo(a,h)anthracene	1.64E-07	1.27E-07	1.3E-07	2.99E-07	5.51E-08	4.07E-08	4.4E-08	1.07E-07
	PCB 180	9.66E-08	7.3E-08	7.45E-08	2.03E-07	3.15E-08	2.06E-08	2.5E-08	6.13E-08
Oral HQ	2,3,7,8-TCDD	1.72E-02	1.68E-02	1.21E-02	3.74E-02	5.82E-03	5.23E-03	3.57E-03	1.22E-02
Inhalation cancer risk	2,3,7,8-TCDD	1.14E-09	2.33E-10	1.12E-09	1.45E-09	-	-	-	-
	Benzo(a)pyrene	3.54E-11	7.54E-12	3.54E-11	4.55E-11	-	-	-	-
	Dibenzo(a,h)anthracene	1.61E-11	3.26E-12	1.63E-11	1.98E-11	-	-	-	-
	PCB 180	7.33E-12	1.17E-12	7.46E-12	8.77E-12	-	-	-	-
Inhalation HQ	2,3,7,8-TCDD	1.67E-04	6.81E-05	1.71E-04	2.67E-04	-	-	-	-

HQ: Hazard Quotient (non-carcinogenic risk), SD: Standard deviation

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In the evaluated scenarios, the hazard quotient (HQ) for 2,3,7,8-TCDD, the only pollutant for which non-cancer reference dosis has been provided, was below the threshold value, set at 1. The highest HQ mean value correspond to food ingestion for farmers ($1.72 \cdot 10^{-2}$). Likewise, the highest cancer risk values were found for the ingestion pathway, mainly due to diet. The mean oral cancer risk values for ingestion ranged between $9.66 \cdot 10^{-8}$ and $6.86 \cdot 10^{-7}$ for the occupational scenario, and between $3.15 \cdot 10^{-8}$ and $2.26 \cdot 10^{-7}$ for the non-occupational scenario. Cancer risk due to inhalation was lower than that derived from the ingestion of pollutants, ranging between $7.33 \cdot 10^{-12}$ and $1.14 \cdot 10^{-9}$ (occupational scenario only). Nevertheless, none of the POPs presented risk values above the threshold (range: 10^{-6}-10^{-4}). Therefore, it can be asserted that the current concentrations of the POPs here evaluated in sewage sludge used for agricultural soil-amending do not mean an important increase in human health risks for both occupationally and non-occupationally exposed populations.

However, POP concentration in sewage sludge may vary in several orders of magnitude. Therefore, not only the application dose, but also the contaminants concentration in sewage sludge and soils, should be taken into account in a comprehensive evaluation. In addition, these facts should be considered in the legislation regarding application of sewage sludge to agricultural soils. The 3rd draft on the working document on sludge (CEC, 2000) defines maximum recommended levels for total PAHs, total PCBs and PCDD/Fs. Compared to the present study, the levels defined by the draft are several orders of magnitude higher than those presented here. Nevertheless, as these are total levels, soil amendment could represent important risks to human health, considering the presence of mixtures of congeners. Because of that, these levels should be investigated in more depth, taking into account the physical-chemical and toxicological properties of each pollutant. As indicated by Schowanek et al. (2004), the need to include certain compounds groups and establish limit values is still a subject under debate, as inside each POP group there are congeners with varied persistence and toxicity.

The uncertainty in the results was less than one order of magnitude. The upper boundary levels (90th percentile) were below the threshold values in most cases. Only

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for 2,3,7,8-TCDD, oral cancer risk for occupational population was slightly above 10^{-6}, but still below the acceptable threshold ranges. This means that, for the present case-study, even considering that the analysis inputs may vary, the accomplished levels of risk are acceptable.

3.5 Sensitivity analysis

A global sensitivity analysis was performed for 2,3,7,8-TCDD, which is the chemical with the highest risk level and presenting the highest variability in the results. The parameters evaluated in the sensitivity analysis were concentration in air (gas and particle phases), soil and plants, and exposure parameters. Scenarios were generated by a Monte-Carlo simulation, with 5,000 iterations. High coefficients of correlation (R^2) were found for the three performed evaluations, indicating that most model input variables act linearly and that higher order interaction effects are not important.

The sensitivity analysis results are shown in Table 5.8. For ingestion values in the occupational scenario, the sensitivity results (R^2=0.78) showed that the main variability in the results was related to the ingestion rate of milk (CR_{milk}) and meat (CR_{beef}). This is due to the fact that the ingestion of meat and milk were found to be the most important pathways in the exposure through diet. The ingestion of meat meant between 57% and 59% of the dietary exposure values, while the ingestion of milk represented between 40% and 42% for all the evaluated scenarios. In consequence, the ingestion of vegetables and the direct ingestion of soil were found as pathways of minor importance. The amount of soil ingested by cattle (Qs_{milk} and Qs_{beef}) are also of pronounced importance This is due to the fact that soil ingestion is the most important pathway for the 2 types of cattle, representing more than 95% of the total ingestion of contaminant. The same tendencies were observed for cancer risk due to ingestion in the non-occupational scenario (R^2=0.75). For inhalation (R^2=0.70), the complexity of the exposure paths was lower and the variability in the results was mainly explained by the exposure frequency (EF), followed by the pollutant concentration in the gas phase.

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Table 5.8. Sensitivity analysis results for 2,3,7,8-TCDD risk values.

	Occupational ingestion cancer			Occupational inhalation cancer			Non-occupational ingestion cancer	
	β value	**S value**		**β value**	**S value**		**β value**	**S value**
CR_{milk}	0.47	0.22	EF	0.65	0.43	CR_{milk}	0.43	0.18
CR_{beef}	0.35	0.12	Level in air	0.40	0.16	Fi_{human}	0.37	0.13
Qs_{beef}	0.32	0.10				Qs_{beef}	0.31	0.10
Qs_{milk}	0.29	0.09				CR_{beef}	0.31	0.10
Fi_{human}	0.25	0.06				Qs_{milk}	0.28	0.08
BW	-0.25	0.06				AFIg	0.24	0.06
AFIg	0.23	0.05				BW	-0.22	0.05

Input parameters definitions and values are shown in Tables 5.1 and 5.2.

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The sensitivity analysis results showed that, for the performed risk analysis, the variability in the results is mainly explained by the variability of the exposure parameters. More accurate results could be obtained with a better knowledge of these parameters. This is a complicated issue as dietary habits significantly change according to the studied area.

The sensitivity analysis also showed the importance of the ingestion rate of soil by cattle. These results suggest that the application of sewage sludge on agricultural soils represents much lower risk values than the amendment of pasture fields. Probably, the substitution of the spreading of sewage sludge by its injection in lower layers could prevent most of the risks related to the ingestion of contaminated soil by cattle. This practice could also prevent occupational exposure through inhalation. Therefore, regulatory efforts should contemplate the peculiarities of each amending site and the types of sludge application to prevent risks to human health, and allow better management practices.

4 Conclusions

In this chapter, the application of sewage sludge on agricultural soils has been shown to be a practice with non-significant human health risks, on the basis of the evaluated conditions. Food ingestion has been proved to be an important exposure route, beingmainly affected by the calculated concentrations in meat and milk. In order to improve the precision of the results, future studies should consider a better estimation of some exposure parameters, with a special emphasis on the dietary pathway. Moreover, the study of other types of soil amendment, such as injection of sludge, should be included. Finally, the insertion of external sources could improve the precision of the results.

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Discussion Part I

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The application of environmental models to predict the fate of POPs in soils amended with sewage sludge allowed evaluating the long-term contamination of these soils. The calculations were performed considering a large time span (between 20 and 30 years). It was proved that all contaminants reach steady-state during this period, and that the time it takes to reach these constant levels is related to each contaminant half life in soil.

The calculation of transport between phases was based on fugacity models. The use of these models provided a good approximation to the reality, with the use of a limited number of differential equations (one for each environmental compartment). It also allowed the calculation of all relevant transport processes, such as volatilization, diffusion and leaching.

Chapter 3 presents a study that evaluated the behaviour of the contaminants and the influence of the soil matrix. For that, virtual contamination levels were applied to two different soils. This work clearly pointed out different trends of accumulation in the soil matrix according to each pollutant group. Most of PAHs and PCBs rapidly degraded, with the exception of benzo(b)fluoranthene. In contrast, PCDD/Fs have been proven to be highly accumulative in soils, with the different accumulation degrees also related to soil half life.

In contrast, no significant differences were found between soil types. However, contaminants with higher removal rates showed a tendency to be more sensitive to soil parameters. This point needed further research as the two evaluated soils are representative of the region of Catalonia, but differences in soils characteristics are expected when the model is extrapolated to other regions.

Chapter 4 was developed based on these results. A sensitivity analysis was performed to quantify and elucidate the influence of input variables and parameters in model predictions.

To perform the sensitivity analysis, the parameters were varied in a wide range through Monte Carlo simulation. In this way, the generated scenarios covered a great amount

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of soil and contaminant characteristics, and their related uncertainty. The results of this study confirmed that half life in soil is the most important parameter of the model. In consequence, any model enhancement depends on a more accurate estimation of this variable.

A variable importance within soil parameters, such as organic carbon fraction, was found. Only for PCB 28, the leaching rate seemed to have some importance in the results. Besides soil half life, the only chemical parameter that presented some importance was K_{OC}, especially for 2,3,4,7,8-PeCDF and PCBs. The high R^2 values proved that the model acts mainly linearly and higher other interaction effects are not important.

Finally, Chapter 5 presented the integration of probabilistic multimedia models (fate, exposure and risk) for the case of sewage sludge amendment on agricultural soils. Four contaminants were selected for the models integration. The study considered current levels of these contaminants in sludge, as well as common application practices, for a time span of 30 years.

The calculated environmental concentrations were similar to those found in the literature. For cancer and non-cancer risk, the likelihood of reaching threshold values is extremely low. In consequence, the application of sewage sludge on agricultural soils does not mean a risk to human health, if pollutants levels in sludge remain. Air inhalation was noted to be an occupational exposure route of minor importance.

The sensitivity analysis showed that exposure parameters were responsible for most of the variability in the results. In this regard, more realistic results could be obtained with more precise information regarding population dietary habits. The amount of soil ingested by cattle is also of pronounced importance. In consequence, a more accurate soil model could give more accurate results. Again, the use of appropriate half lives in soil is crucial to improve the obtained results.

Finally, it is important to highlight that models are an approximation of the reality, and they have some limitations. The representation of the complexity of environmental

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systems in a changing world is a difficult task. Due to this fact, some issues are crucial for better modelling practices. First, models objectives must be properly defined. Then, the developed models must be evaluated, tested and checked. Finally, models must be transparent and their assumptions clearly stated. The environmental models reported in this thesis have been developed considering all these factors. However, the field of model development and evaluation still have some challenges to be solved, such as the representation of highly complex systems and the improvement of uncertainty management techniques. The use of good modelling practices in this work will ease future improvements, when necessary.

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Part II

Decision models

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Chapter 6

A spatial multicriteria decision analysis to manage sewage sludge application on agricultural soils

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1 Introduction

Due to environmental and economic costs, the management of an increasing production of residues (e.g., sewage sludge) is a current issue of great interest. In recent years, an increased production of this residue has been experimented in a series of countries.

Nowadays, the main criteria for sewage sludge management are legislation and economic costs. Metal levels in sewage sludge and agricultural soils are defined in the European legislation (CEC, 1986). However, other issues such as potential impacts to humans and ecosystem, including human exposure to organic contaminants through different pathways are not addressed. Another point of concern is that amending soils with sewage sludge may lead to surface water and groundwater contamination, as a result of pollutants movement through the soil matrix. Besides that, economic issues such as the managing cost (function of the amount of sludge produced and the treatment process) and the transport cost (according to the distance between the wastewater treatment plants and the agricultural field) must be also taken into account.

The purpose of this chapter is to present a Spatial Multicriteria Decision Analysis (SMCA) to define the optimal allocation for amending agricultural soils with sewage sludge. For that, the problem of disposing sewage sludge is presented and novel decision support methodologies are described. SMCA allows assessing the impacts and the benefits of the different disposal alternatives. It involves a set of geographically defined alternatives (events) from which a choice of one or more alternatives is made with respect to a given set of evaluation criteria.

2 Sewage sludge management

Sewage sludge is the main residue of Wastewater Treatment Plants (WWTP). It includes the sludge originated in treatment plants of domestic wastewater that is occasionally mixed with industrial wastewater and/or run-off rain water. Sewage sludge properties depend on wastewater pollution load, and technical characteristics of the treatment

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plant. Due to water treatment, the pollution contained in water is concentrated and also some chemicals are transformed during the treatment process (Katsoyiannis and Samara, 2005). Some of these compounds, such as organic matter and nutrients, may be usefully reused. However, sewage sludge also presents some pollutants, as heavy metals (Aubain et al., 2001; Metcalf and Eddy, 2003), Persistent Organic Pollutants (POPs) (Eljarrat et al., 2003; Harrison et al., 2006) and pathogens (Aubain et al., 2001; Metcalf and Eddy, 2003). Recent studies also point out the presence of pharmaceuticals in this residue (Radjenovic et al., 2009) as a consequence of the extensive use of these substances by the population. For this reason, sewage sludge must be properly disposed to avoid risks to humans and ecosystems.

During the last few decades, the increase of sludge production, as a consequence of industrial development, population increase and amplification of treatment services, has become an environmental problem for several countries. Once treated, sludge can be recycled or disposed using one of the following 3 routes: application on agriculture, incineration (combustion) or landfilling. The valorisation of this residue as an alternative energetic source in cement plants is nowadays a recycling route of increasing interest. Other disposal routes are land reclamation, as well as other combustion technologies including wet oxidation, pyrolysis and gasification (Fytili and Zabaniotou, 2008). Each of these routes has different economical costs and environmental impacts. The selection of an option on a local basis reflects local or national, cultural, historical, geographical, legal, political and economic circumstances (Fytili and Zabaniotou, 2008).

Among these routes, soil amendment with sewage sludge is the preferred alternative. However, studies to elucidate the benefits of this practice to the population are still needed. Governments are pressured by the population because of the risks related to food safety that this practice may represent. Therefore, existing regulations and further controls to minimize human exposure and derived risks must be elaborated.

Regarding the use on agricultural soils, stakeholders may have different opinions. Farmers are motivated by the low cost that represent the use of sewage sludge as a

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fertilizer. However, as food industries have specific quality requirements, farmers need some guarantees related to sludge quality to prevent a reduction in their market share. Landowners are concerned about a possible decrease in land value. On the other hand, environmental agencies are interested in the disposal of this residue with a low economical cost, but assuring that this practice does not represent a threat for the population. In most cases, administrators have implemented policies to regulate sewage sludge application on soils, which is considered the best environmental and economic option (Aubain et al., 2001, 2002). However, the population still needs more information concerning the risks and benefits related to this management practice.

With appropriate use, the application of sewage sludge on agricultural fields has benefits to soils and crops. This amending practice improves soil fertility, reduces the use of chemical fertilizers, and it is a relatively inexpensive solution. It also improves the soil structure by the addition of Organic Matter (OM) to the soil matrix and nutrients recirculation through soil (Schowanek et al., 2004). Other benefit of sludge application is the reduction of the potential for surface runoff and water erosion.

OM degradation increases the soil content in compounds of agricultural value (such as N), which are slowlier released than in the case of mineral fertilizers, making them available to crop for a longer period. When compared to other types of fertilizer, such as urban compost and animal manure, sewage sludge has higher OM concentration, especially the composted sludge. Furthermore, the OM added to soil through composted sludge is more stable than for other types of sludge. In this case, organic matter mineralizes slowlier, and nutrients are released at a slower pace, reducing the potential risk of nitrogen leaching to groundwater.

Nitrogen is mostly found under organic form in sludge. The value of this residue as a fertilizer is determined by the capacity of organic N to be mineralized. The nitrogen availability is influenced by the type of sludge, temperature, humidity, pH and soil texture, as well as the application conditions (Aubain et al., 2001). The content of N in sludge may vary depending on the sludge treatment type, operational and storage conditions (Mantovi et al., 2005).

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Although the application of sewage sludge on soils represents a number of benefits to soil structure, the presence of contaminants in sewage sludge may result in risks to humans and ecosystems. This contaminant movement among environmental compartments may lead to soil contamination, and indirect emissions to air and water. Various pollutants are present in sludge, being heavy metals and persistent organic pollutants (POPs) two of the chemical groups of most concern.

The presence of heavy metals in soil has been reported in the literature (Fytili & Zabaniotou, 2008; Nadal et al., 2004a, 2007). These elements are naturally present in different concentrations in background soils, due to local geological characteristics of the field. They may also originate from anthropogenic sources such as fertilizers, animal manure, sludge application, or atmospheric deposition.

Regarding sludge application, these chemicals are distributed between different soil media, tending to accumulate in the upper layers of the soil, attached to organic or mineral particles. Among soil parameters, pH is the most important factor in metals mobility and bioavailability. Heavy metals runoff may lead to surface water contamination.

POPs have been found in soils contaminated by different sources (Nadal et al., 2004b, 2007, 2009a; Wyrzykowska et al., 2007). POPs are toxic and persistent chemicals, characterized by their bioaccumulation potential and long-range transport capacity. These pollutants move through different environmental compartments. Within these fate processes, POP concentration dynamically changes over time and leads to accumulation in the food chain. As a final step, these contaminants may be transferred to human beings. The most important factors in POP mobility and degradation in soil are their half-life in soil and octanol-water (K_{ow}) partition coefficient (Passuello et al., 2009). In addition, soil OM content and octanol-carbon (K_{oc}) partition coefficient are expected to influence also POP fate on soils.

As a result of amending soils with sewage sludge, humans are exposed to heavy metals and POPs through ingestion of soil, plant and meat, particle and air inhalation

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(Passuello et al., 2010). The exposure to these contaminants may therefore lead to human health risks.

The most relevant legislation concerning sludge application on soils is the Council Directive 86/278/EEC (CEC, 1986), which establishes a limit for concentrations of heavy metals in soils and sludge. Among the European Community members, six of them (Greece, Ireland, Italy, Luxembourg, Portugal and Spain) have implemented limit values similar to the Directive, while eight countries (Belgium, Denmark, Finland, the Netherlands, Sweden, Estonia, Latvia and Poland) have applied more restrictive limit values. Moreover, some regulations include threshold values for pathogens (France, Italy and Luxembourg) and organic compounds (Austria, Belgium, Denmark, France, Germany and Sweden), which are not included in the Directive.

The European Community is aware of the necessity of including organic contaminants in regulations. So far, the 3rd draft of the working document in sludge (CEC, 2000) defines threshold values for concentrations of organic compounds in sludge for use on land. This draft also establishes limit values for concentration of heavy metals in soil and sewage sludge, which are more restrictive than those previously defined by the Council Directive (CEC, 1986). The Council Directive 91/676/EEC (CEC, 1991a), related to the protection of waters against pollution caused by nitrates from agricultural sources, is also of concern, as this legislation requires identification by Member States of Nitrates Vulnerable Zones. Very few regulations specifically address the use of sludge in routes other than the recycling in agriculture (use in silviculture, green areas and in land reclamation). Only the use of sludge on forest soil is mentioned by some national regulation.

Regarding economic price, sewage sludge management represents internal and external costs and benefits (Aubain et al., 2002). Internal costs comprise of: investment, transport, management, storage and application costs. In turn, internal benefits represent the profits obtained due to the substitution of a commercial fertilizer. Söderholm and Sundqvist (2003) defined externality as ‘an unpriced benefit or cost directly bestowed or imposed upon one agent by the actions of another agent.’

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In this sense, the external costs and benefits include social, environmental and human health impacts.

The benefit of substituting a regular fertilizer by sewage sludge is relatively easy to estimate, taking into account field and crop characteristics. In addition, some methodologies, such as that developed by Koo and O'Connell (2006) to estimate the agricultural productivity depending on the type of crop and land use alternatives, may be applied. This agronomic model is based on nitrogen response curves and the crops' price. The yield (in t ha^{-1} $year^{-1}$) is a function of nitrogen application (kg N ha^{-1} $year^{-1}$), and the price is estimated by the crops market value (€ t^{-1}).

The transport and application costs vary according to the solids content of sludge and the type of crop. Transport costs are also related to the distance between the agricultural field and the WWTP. For this reason, for WWTP situated in large cities without agricultural fields in the vicinities, this practice may be too expensive and other alternatives should also be studied. Other parameters of interest are the amount of sludge produced and the nitrogen content of sludge, which is related to the application dose.

The external cost and benefits are normally more complicated to measure than the internal costs and benefits, as they involve social and environmental costs, for which no market prices apply. Besides, as there is a lack of an established methodology to determine these risks in monetary value, the results may present a high subjectivity. Instead of that, the use of other types of evaluation, such as Multicriteria Decision Analysis (MCDA), is highly recommended.

3 Multicriteria Decision Analysis

Sewage sludge management is a complex decision problem, due to the different objectives of the involved stakeholders. There are social aspects involved, such as food safety and related human health risks. Moreover, the management must also be economically viable. The most suitable agricultural areas should be selected based on all the previously cited issues.

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To define the suitability of each agricultural area to receive sewage sludge, three main attributes or criteria must be considered: economic, social and environmental. These criteria comprise several subcriteria of this managing practice. For example, the economic criterion must take into account transport, storage and application costs. The environmental criterion evaluates soil contamination aspects, including soil and landscape parameters. These subcriteria may not be directly integrated, as they are not measured on a common basis, and some of them are incommensurate. Due to this fact, a MCDA must be developed to solve this problem.

A criterion is a tool constructed for evaluating and comparing potential actions according to a point of view which must be (as far as it is possible) well defined. This evaluation must contemplate, for each action, all the pertinent effects or attributes linked to the considered opinions (Roy, 2005). The use of MCDA allows dealing with a broad spectrum of points of view, through the construction of a criteria family that preserves the original meaning of the corresponding evaluation. Also, MCDA eases the debate between stakeholders, thanks to its clear structure, facilitating the evaluation of the rules (weights and performing-scoring scales) on the decision process. This is a key fact if we consider that the analysis results depend on weights and utility scales together.

The methodology is based on the hypothesis of a rational and consistent decision maker (Bridges et al., 2004), and it implies the existence of both value functions and suitable aggregation operators (Critto el al., 2006). As a consequence, a key point in MCDA is the criteria selection, which is performed by experts. The number of criteria must be sufficient to consider all the relevant issues of the problem, and minimal enough to avoid redundancy. Also, the criteria attributes must be decomposable to simplify the process.

There are a number of MCDA procedures reported in the literature. The selection of the best procedure for each case-study is still a subjective issue. The most used methods are: (i) outranking methods, such as ELECTRE (Figueira et al., 2005), PROMETHEE (Brans and Mareschal, 2005); and (ii) Multiatribute Utility Theory (MAUT) methods,

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such as weight summation methods (Torra and Narukawa, 2007) and Analytic Hierarchy Process (AHP) (Saaty, 1980; Ramanathan, 2001; Saaty, 2005).

3.1 Geographical Information Systems and Multicriteria Decision Analysis

Sewage sludge management is based on several attributes. Many of these are related to the landscape properties and the location of each agricultural site. For example, pollution in rivers due to metals runoff is correlated with rainfall, terrain slope and proximity to open waters. All these parameters may be evaluated in a more global way with the use of Geographic Information Systems (GIS). The use of GIS for decision making allows evaluating and grouping a large amount of spatial data. In consequence, modellers may consider a large number of feasible alternatives to be evaluated on the basis of multiple criteria (Malczewski, 1999).

In recent years, GIS have emerged as a very important tool for land use suitability analysis. GIS can recognize, correlate and analyse the spatial relationship between mapped phenomena, thereby enabling managers to link different sources of information, perform sophisticated analysis, visualise trends, project outcomes and strategise long-term planning goals (Sumathi et al., 2008; Behzadian et al., 2010). Due to these characteristics, GIS have been largely employed to solve spatial decision problems. These may be defined as those problems in which the decision implies the selection among several potential alternatives that are associated with some specific geographical locations. Spatial Multicriteria Decision Analysis (SMCA) refers to the application of MCDA to manage spatial problems. According to Malczewski (2004), two considerations are of critical importance for SMCA: (i) the GIS capabilities of data acquisition, storage, retrieval, manipulation, and analysis; and (ii) the models capabilities for combining the geographical data and the decision maker preferences into unidimensional values of alternative decisions.

As in any multicriteria decision model, several operators may be applied to solve spatial decision problems. Boroushaki and Malczewski (2008) proposed the integration of GIS in an extension of the Analytic Hierarchy Process (AHP) using the quantifier-guided Ordered Weighted Averaging (OWA) procedure. Chakhar and Mousseau (2008)

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suggested a framework to facilitate the incorporation and use of outranking methods in GIS. Geneletti (2007) proposed a methodology based on commonly available data to assess the nature conservation value of agricultural landscapes, and to generate cartographic results to be used as decision variables in planning. Cases of SMCA by using Internet and webmapping techniques have been described (von Haaren and Warren-Kretzschmar, 2006). Agriculture is one of the major application areas of SMCA (Malczewski, 2006). In recent studies, Ceballos-Silva and Lopez-Blanco (2003) applied SMCA to identify suitable areas for the production of maize and potato crops. In turn, Morari et al. (2004) applied SMCA for selecting criteria of best management practices in agricultural sites.

4 Spatial Multicriteria Decision Analysis implementation

As stated before, there are several MCDA methods reported in the literature that can be applied as part of the SMCA. The revision of all the existing methods is not the scope of this work, so we have selected three methodologies that fit well with the decision problem presented. The first part of this section explains briefly the weighted summation methods and the AHP, which are the most used methods for SMCA (Malczewski, 2006). Then, fuzzy methods are introduced. These methods have emerged in the last few years to deal with uncertainty in environmental management. Although the number of publications regarding the application of this method is still limited, an increasing tendency is expected due to the possibility of dealing with uncertainty in environmental modelling.

4.1 Weighted Summation Methods

Weight summation methods are the most simple and applied methods in SMCA. For each alternative and criterion, a score is computed. This score is defined as the weighted sum of its evaluations. Among the existing weighted summation methods, the ordered weighted averaging (OWA) operator comprises the weights of relative criterion importance and order weights. The OWA operator was introduced by Yager (1988) and provides a parameterized family of aggregation operators that includes the arithmetic mean, the maximum and the minimum.

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Another method that has been applied to the GIS platform is the Logic Scoring of Preference (LSP). LSP method is based on mathematical models that use Generalized Conjunction/Disjunction (GCD) and other continuous preference logic functions (Dujmovic, 2007). LSP operators take into consideration the different levels in the hierarchy of criteria and user weights and constraints over those criteria. The aggregation of partial preferences is based on GCD logic that is used for modelling simultaneity and replaceability (Dujmovic and Larsen, 2007).

These approaches are GIS-implemented using algebraic operations and cartographic modelling. The relative simplicity of these methods makes them easily understandable by decision-makers. Despite this, sometimes these methods are applied without a full understanding of the assumptions underlying this approach. Steele et al. (2009) pointed out that the numerical criteria weights must reflect the relative importance of the criteria. For that, the weights must consider the way in which the performance scoring scales for the criteria have been calibrated. This means that besides a good weighting election, the decision maker must be aware of the importance of the scoring scales to avoid an overvaluation of some criteria.

4.2 Analytic Hierarchy Process

The Analytic Hierarchy Process (AHP) (Saaty, 1980) is a method for analyzing complex decision problems under multiple criteria. The AHP is a theory of relative measurement on absolute scales of both tangible and intangible criteria. This theory is based on both judgment of expert people and existing measurements needed to make a decision (Saaty, 2005). The management options for a particular decision problem are characterized by their attributes with respect to a set of detailed criteria.

The AHP procedure involves three major steps: (i) development of the attributes hierarchy, (ii) pairwise comparison of the hierarchical structure attributes, and (iii) construction of an overall priority rating (Boroushaki and Malczewski, 2008). According to Malczewski (2004), AHP can be used in two different ways in a GIS environment. The first approach consists on deriving weights associated with an attribute map layer. The

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second approach uses the AHP principle to aggregate the attributes of all hierarchy levels.

A drawback of the AHP method is the use of redundant judgments for checking consistency, which can exponentially increase the number of judgments to be elicited from decision makers (Ramanathan, 2001). For example, to develop the pairwise comparison matrix of 6 criteria, 15 judgments are needed. For the same analysis, if we have N alternatives, the number of judgments would be 15·N. For this reason, AHP is habitually used to compare a small number of alternatives concerning the overall goal (Wang et al., 2009), as the decision makers effort may be too large in more complex problems.

Despite the widespread use of AHP, some researchers question the theoretical functions of the method (Malczewski, 2004). The implementation of this approach also requires a complex data elicitation process (Kumar et al., 2007). Another problematic issue of this method is the rank reversal problem. Several studies state that the ranking of alternatives may be altered by the addition of another alternative (Ramanathan, 2001). Lootsma (1999) found that the alternatives ranking was different when using AHP with ordinal (ranking) and cardinal (scores) information. However, Saaty (2005) stated that the ranking is preserved any time another alternative is created or removed from the analysis, if an ideal alternative is created (ideal/reference point methods).

4.3 Fuzzy knowledge-based models

Fuzzy knowledge-based models have been largely applied in environmental modelling in recent years (Ocampo-Duque, 2008). These methods are based on a natural language that is intuitive for most of the decision makers, as they use rules in the form IF…AND…THEN. For instance, “IF” vegetation cover is low “AND” rainfall is high, “THEN” the possibility of erosion is high. This method has been developed for modelling complex systems in uncertain and imprecise environments (Ross, 2004), and it has contributed to the challenge of interpretation and translation of natural language

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specifications into formal mathematical expressions (Boroushaki and Malczewski, 2008).

As in any decision making method, the development of a fuzzy knowledge-based model requires the determination of model structure, criteria, subcriteria and connections between them. Once the rules are selected, a fuzzy inference method is used to process this knowledge and compute output values. The input values can be numerical, linguistic or fuzzy sets. This is an advantage, as it allows dealing with uncertain information.

Fuzzy systems have been recently employed to support decisions in environmental management. Ocampo-Duque et al. (2007) applied a hybrid approach, based on fuzzy inference systems and artificial neural networks to classify the ecological status of surface waters. The combined application of fuzzy systems and GIS has not received the same attention as the weighting summation approaches. However, some interesting works have emerged recently. Reshmidevi et al. (2009) presented a GIS-integrated fuzzy rule-based inference system to evaluate land suitability in agricultural watersheds. Pathak et al. (2008) applied a fuzzy optimization method in a GIS environment to evaluate groundwater vulnerability to pollution. Chang et al. (2008) integrated a fuzzy multicriteria decision-making tool in a GIS platform to define the best area to locate a landfill in a fast-growing urban region and to define a fund distribution for a municipal incinerator using a mixed approach, integrating GIS and fuzzy AHP (Chang et al., 2009). The method proved to be an effective way to describe the environmental impacts in the context of decision analysis as well as to identify the associated weights for fund distribution.

5 General Framework

In this section, the guidelines to develop a SMCA are presented. An introductory example is given to illustrate method application and improve the understanding of the tool. For a correct model application, this example should be extended to fit the decision objectives for each specific case.

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The decision making process has three main parts (Figure 6.1): evaluation, elaboration and integration. In the evaluation phase, the decision problem is defined and recognized. The decision makers must explore the available legislation, current management practices and tendencies related to the decision problem. The criteria are also defined in this step. In the elaboration phase, the selected criteria are translated into preferences, and the SMCA methodology to be applied is defined. Finally, in the integration phase, the previously selected data is modelled, the results are evaluated and a sensitivity analysis is performed.

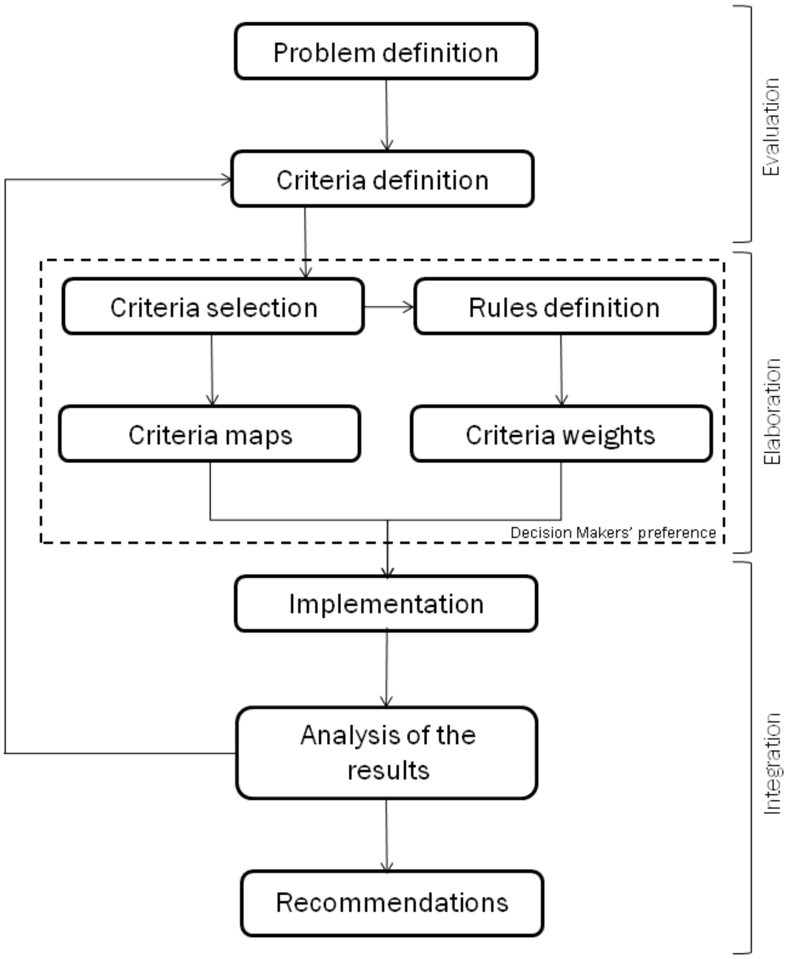

Figure 6.1. Framework for SMCA

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5.1 Evaluation Phase

Sewage sludge management is a complex decision problem that involves the opinion of different groups of stakeholders. This particular SMCA is developed based on the stakeholders' pressures and opinions, as well as the type of information and knowledge that can be acquired. The SMCA proposed here takes into account environmental, social and economic criteria. Environmental criterion comprises soil characteristics and landscape properties of the agricultural area. The social criterion is represented by population characteristics while the economic criterion considers sludge production and WWTP characteristics.

Recognizing the problem also means selecting the most appropriate criteria maps to perform the evaluation. To select the most appropriated SMCA tool, the decision maker needs to consider all the problem characteristics and constraints, and to analyse the available information.

The use of SMCA for sludge management has the objective of defining the optimal allocation for this residue in agricultural areas, satisfying environmental, economic and social criteria. Each criterion is divided in subcriteria to perform the evaluation. For example, the environmental criterion has the subcriteria 'risks of contaminating water' and 'risks of contaminating soils'. The 'risks of contaminating soil' comprise soil attributes (such as pH and OM content) and sewage sludge attributes (contamination levels, type of treatment, etc). The environmental criterion also includes the possible alterations in soil biodiversity and productivity.

Amending soils with sewage sludge may lead to crop contamination. Many of the contaminants that are found in sludge are persistent, may accumulate in the food chain and are potentially harmful to humans. This is an environmental aspect of the problem. A criteria hierarchy for this problem is proposed in Table 6.1.

Table 6.1 presents a brief list of criteria that may be considered in such an evaluation. Depending on the agricultural site, more criteria could be added or some of the proposed criteria could be excluded. The selected maps must be treated to evaluate

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the selected criteria. For example, the performance of the sub criterion “Groundwater (GW) contamination” is related to the attributes: soil classification, terrain slope, aquifer depth, aquifer recharge, temperature and rainfall. For instance, a coarse soil (sand) has a higher risk of GW contamination than a fine soil (clay). Geographical areas with high slope and high rainfall are likely to transport pollutants by run-off, tending to contaminate surface waters instead of GW.

Table 6.1. Criteria hierarchy of the proposed problem

Criteria	Subcriteria
Environmental	Soil Contamination
	Surface Water Contamination
	Groundwater Contamination
Human Health	Human Environmental Exposure
	Dietary Exposure
Economic	Transport Costs
	Storage Costs
	Application Costs

In general terms, administrations have good mapped information related to agricultural areas. The terrain characteristics, as slope, landscape temperature, soil classification, and crop type are relatively easy to find in public databases. Nevertheless, sometimes these information sources are not as detailed as it would be necessary to assure a good decision making. Another problem of data mining is the need of specific environmental data, as much as levels of micropollutants (heavy metals, POPs, etc.). Some of the data may be found at a continental level, whereas some data may be found in a more regional level. The selected data should have, as far as possible, the same accuracy level. Finally, the procedures for selecting the attributes should be based on the desirable properties of these attributes (Malczewski, 1999).

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5.2 Elaboration phase

After selecting the attributes, the decision makers must define the weights and the preference scale of each attribute. This step is called the elaboration phase. The models selection must consider the existence of interrelations between the selected criteria. The preference scales are characterized depending on available maps and data quality. To the best of our knowledge, there is no established method to define the preference scales.

To better understand the problem elaboration, let's consider the previously cited subcriterion "risks of contaminating soils". For this criterion, the following hierarchy is suggested (Table 6.2).

Table 6.2. Example of Criteria Hierarchy and Maps Selection of the Proposed Problem

Criterion	Sub criterion	Parameters (maps)
Soil Contamination	Effects on Soil Structure	Soil Parameters: OM, pH, Texture, Pollutants Concentration

The method used in maps aggregation is determined by the available information and must reflect the decision makers' preferences. For example, when the available data is deterministic, a weight summation method may be applied. However, if the inputs are probabilistic or the decision maker wants to insert uncertainty in the evaluations, a method capable of dealing with uncertainty must be selected.

To perform the evaluation, each input map must be translated on a 'preference map'. This means that for each attribute map, the decision makers must define utility values. For example, it is desirable to have a basic soil to apply sewage sludge because the metals availability is lower in this type of soil. As a consequence, the decision makers define that a pH of 8 or higher has a high utility for sewage sludge application. In contrast, a pH that is lower than 6 has the worst utility for this attribute (see Figure 6.2). The preference scale values between 6 and 8 are linearly represented. Utility values are between zero and one. For each pixel, the sampled pH values are translated

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to a utility value, generating the preference map. This procedure must be performed for each input map. A simple way of defining criteria weights and preference scales is performing series of interviews with the stakeholders, managers, experts from the environmental agency and research centres.

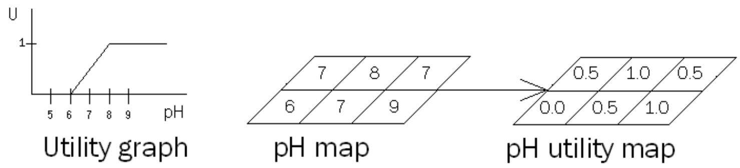

Figure 6.2. Building Utility Maps

Once all the utility maps have been created, these maps are integrated using the selected method. Figure 6.3 shows the maps hierarchy in this particular example.

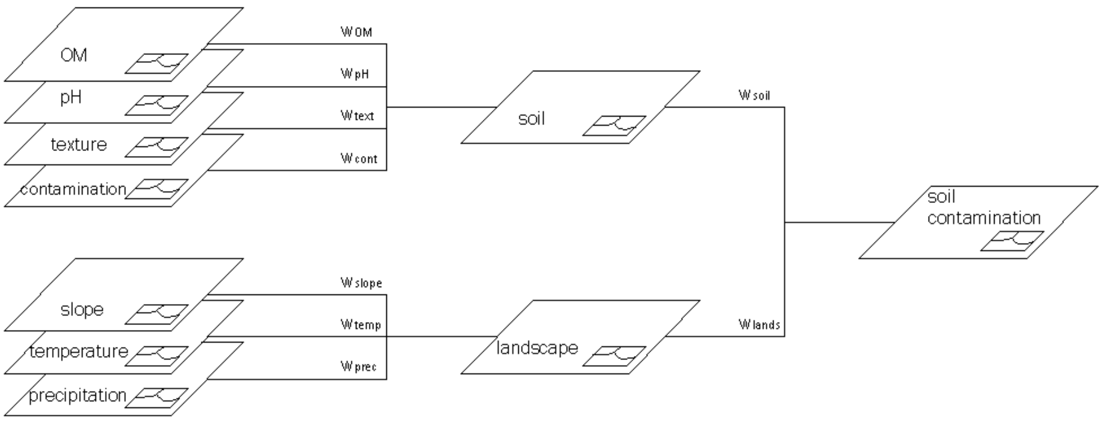

Figure 6.3. Chart of the Integrated System

5.3 **Integration Phase**

Having elaborated the decision problem, the models are implemented. The acquired knowledge is structured and the results are presented in a way that experts can easily understand and contribute with their opinions. In our example, the integration is performed in two steps (Figure 6.3). Firstly, all the attributes related to soil characteristics are integrated in a map called 'Soil'. At the same level, landscape

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characteristics are integrated in a map called 'Landscape'. The integration procedure considers the aggregator and the weights previously defined. Then, the two generated maps are integrated in a map called 'Soil Contamination'. This map reflects the suitability of the studied areas to receive sewage sludge as an amendment, considering the 'Soil Contamination' subcriterion. This step is repeated for all the defined attributes.

In this part, it is also important to clearly show the model assumptions and the restrictions on the application of the decision system. This practice allows validating the model by confronting the results with the stakeholders' experience. In this way, the rules, utilities scales and weights may be refined, adjusted, corrected or extended. Besides, the model stability must be checked. A good system evaluation passes through a Sensitivity analysis (SA). A SA evaluates how the uncertainty in the output of a model can be apportioned to different sources of uncertainty in the model input. In general, global SA methods provide better information as they evaluate globally all the input parameters, in contrast with local or one-factor-at-a-time methods (Saltelli, 2005).

Another way of evaluating the models is through a robustness analysis, which studies the stability of the ranking with respect to the most important elements in the objective and subjective data. Robustness analysis evaluates how the changes in model inputs and structure may lead to changes in the alternatives ranking.

All these techniques may also deal with uncertainty management. Uncertainty is inherently present in all methods that try to reproduce real-life systems, and is normally related to lack of information, subjective information (human judgment) and partial knowledge about the system. The previously cited fuzzy logic based models are a major approach to represent uncertainty in MCDA.

6 Conclusions

This chapter describes the main issues of sewage sludge management, considering its application on agricultural soils. A brief example of SMCA implementation, through an

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easily understandable system, is also provided. In the case of sewage sludge management, the method may clearly point out the best areas to amend with sewage sludge. Defining which method and which criteria must be selected is still a subjective issue. In many cases, models selection depend more on experts experience than on stakeholders expectations. This is a problematic issue as the decision makers find hard to validate systems with high complexity. That is the reason why the performance of a SA is so important. In the field area of SMCA, this point needs further efforts in order to improve spatial decision making.

Another problem in SMCA is coupling spatial information with human reasoning. People in general do not readily understand some geographical concepts, especially when dealing with uncertain information as temporal series on a map. Systems that lead with uncertainty, could improve people's capacity in understanding this type of problem.

Finally, the design of a decision support tool must consider the final user. Data treatment and system representation depends on the use of this tool. If the system is designed for external use, it must have a friendly interface, allowing the final user to easily change the key parameters.

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Chapter 7

Development of the spatial decision making tool

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UNIVERSITAT ROVIRA I VIRGILI
DEVELOPMENT OF ENVIRONMENTAL TOOLS FOR THE MANAGEMENT OF SEWAGE SLUDGE ON AGRICULTURAL SOILS
Ana Carolina Passuello
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1 Introduction

In recent years, geographic information systems (GIS) have been extensively applied in several environmental fields such as vulnerability assessment (Kattaa et al., 2010), human health (Nadal et al., 2006, Poggio and Vrscaj, 2009) and ecological risks determination (Schriever and Liess, 2007). Another area of increased use of GIS platform is environmental decision support systems (Passuello et al., 2011).

Spatial multicriteria decision analysis (SMCA) refers to the application of MCDA tools in a GIS platform to solve spatial decision problems, whose decision implies the selection among several potential alternatives that are associated with geographical locations. Two considerations are important to solve spatial decision problems: (i) GIS capabilities of data acquisition, storage, retrieval, manipulation and analysis, and (ii) models capabilities of combining the geographical data and the decision maker's preferences into unidimensional values of alternative decisions (Malczewski, 2004).

Several examples of SMCA application in environmental management are reported in the literature. Agriculture is one of the major application areas of SMCA (Malczewski, 2006). For instance, Ceballos-Silva and Lopez-Blanco (2003) applied SMCA to identify suitable areas for the production of maize and potato crops. Morari et al. (2004) applied SMCA for selecting criteria of best management practices in agricultural sites. However, to the best of our knowledge, the management of sewage sludge on agricultural soils has not yet been studied through spatial analysis.

In addition, there is a lack of SMCA studies that deal with the variability and uncertainty inherent in environmental models. Sensitivity analysis (SA) is crucial in this type of models, due to the high subjectivity related to the selection of criteria and model rules. Among the existing methods, global sensitivity analysis (GSA) has been applied successfully in spatial decision making. For instance, Gomez-Delgado and Tarantola (2006) applied GSA to a case-study to define the best location for a hazardous waste landfill site. In turn, Crosetto and Tarantola (2001) applied GSA to a hydrologic model to provide real-time flood forecasting.

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The exploratory method is another method successfully applied in spatial decision making. Store and Kangas (2001) used the exploratory method to validate their model and to identify the habitat-suitability maps for one forest specie. Moffett et al. (2006) applied the same method in a model that defined priority areas for biodiversity conservation. Valente and Vettorazzi (2009) made a comparison between three different sensitivity analysis methods: Monte Carlo simulation, the exploratory method and the AHP-OWA module, and concluded that the exploratory method is a good blend to perform sensitivity analysis in multicriteria evaluation.

In this chapter, the development of a spatial multicriteria decision model to indicate the best areas to amend with sewage sludge in Catalonia (NE of Spain) is proposed. Results evaluation tools were applied to provide a better understanding of the model. The model evaluation was performed through two different methods: the global sensitivity analysis and the exploratory method.

2 Materials and methods

Complex spatial decision problems are typically ill-defined (Feick and Hall, 2004). For this reason, a clear definition of the framework for the assessment of the decision problem must be performed at the beginning of the evaluation. Figure 7.1 shows the main steps of the analysis. First, the problem must be precisely characterized and the objectives determined. The next point is the selection of the alternatives and criteria definition. In this step, the type of data needed for the study is specified and the maps are elaborated or treated. Then, the alternatives and criteria are crossed and the decision rules and the criteria weights are elaborated and applied. At the end of the analysis, the results are evaluated and the model is adjusted when necessary. In this chapter, we propose the application of two methods for model evaluation: the global sensitivity analysis and the exploratory method.

Decision making deals with multiple groups with non-complementary and often conflicting objectives, therefore clearness is essential during the decision making process. Sensitivity analysis (SA) procedures are of great help on models evaluation and calibration. SA can help to reduce uncertainty in how a MCDA method operates

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and to determine the stability of its outputs by illustrating the impact of introducing changes to specific input parameters (Crosetto et al., 2000). In the following point, we briefly explain the tools selected for this study.

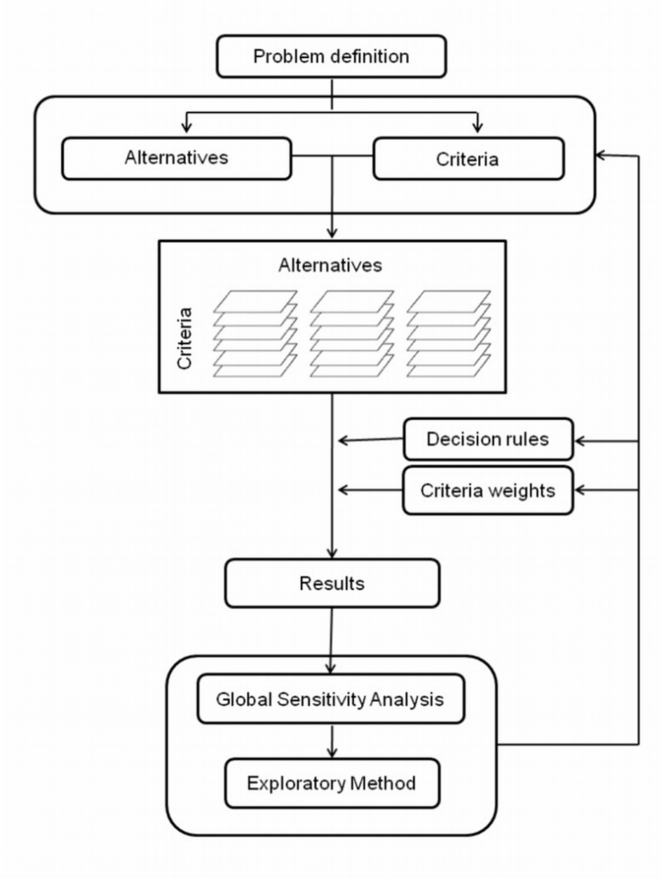

Figure 7.1. Framework for SMCA.

2.1 Sensitivity Analysis

Due to the high complexity of the environmental systems, environmental decision making is a complicated issue. In addition, there is large number of actors or stakeholders that are involved in the decision problem, and a large number of potential management options. These factors in combination may lead to a high subjectivity in the decision process.

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Good modelling practice requests that the modeller provides an evaluation of the confidence of the model, which can be achieved by assessing the uncertainties associated with the outcome (response) of the model itself (Crosetto et al., 2000). The application of sensitivity analysis tools in decision support systems allows evaluating and checking model appropriateness to the problem. SA results are useful for the experts to better understand the model outcomes.

Several methods to perform sensitivity analysis have been described in the literature (Saltelli et al., 2004). However, the application of these methods in spatial decision making is still limited. In this chapter, the application of two methods is proposed. Firstly, the global sensitivity analysis (GSA) was applied to identify the most sensitive input parameters of the model. Then, considering the generated ranking of the most sensitive maps, the exploratory method was employed to determine the error related to the extraction of these maps from the analysis. While GSA considered the variability of the whole region, the exploratory method assessed the error apportioned in each of the evaluated pixels.

2.1.1 Global Sensitivity Analysis

Global sensitivity analysis (GSA) methods study how the uncertainty in the output of a model can be apportioned to different sources of uncertainty in the model input (Campolongo et al., 2000). Contrarily to local approaches, GSA methods consider the full range of uncertainty of the inputs, which are varied simultaneously (Lilburne and Tarantola, 2009). The method also allows the user to quantify the importance of groups of inputs.

GSA is a variance-based method that has the capability to compute sensitivity indices regardless of the linearity or monotonicity, or other generic assumptions on the underlying model (Lilburne and Tarantola, 2009). Depending on the adopted setting, GSA techniques provide first-order and/or higher-order sensitivity measures, including total effects (Saltelli et al., 2004). First-order sensitivity measures are considered suitable for linear models. Higher-order sensitivity indices, responsible for interaction

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effects among input factors, are usually not estimated because of their high computational cost (Gomez-Delgado and Tarantola, 2006).

Among GSA techniques, standardized regression coefficients (β values) may be applied when the inputs are independent. This method is sensitive to all input distributions, and provides a decomposition of the output variance according to the input factors. Campolongo et al. (2000) stated that estimators such as the standardized regression coefficients have a limit of being as good as the regression on which they are based. Because of that, the model coefficient of determination (R^2) should always be determined to give an indication of the model behaviour (linearity, monoticity). In this work, standardized regression coefficients were calculated by means of the sensitivity analysis library of the software Analytica (Lumina, 2008).

2.2 Exploratory Method

Exploratory modelling attempts to capture major theoretical uncertainties in a set of model runs that investigate the implications of different parameterizations to explore the magnitude of possible changes across the model (Pennington, 2007). Within the method, different scenarios are performed. In this study, the scenarios were selected based on the results of the previous step (GSA), where the most important maps were defined. Each scenario corresponds to the alteration of the weights of one input map. The corresponding weight of this map is set to zero, and the other weights are proportionally redistributed in each level (Valente and Vettorazzi, 2009).

Pennington (2007) stated that the goal of this evaluation is not predicting a real system, but improving the understanding of how specific changes impact the results of the model. The generation of scenarios may be a laborious work, especially when the number of inputs is high. In this case, the previous application of GSA will allow evaluating the most important parameters, and specific scenarios may be performed in this step.

In contrast to GSA, the exploratory method evaluates the changes in each pixel of the territory. This method allows quantifying the number of pixels that have changed values

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in their outputs when the weights are changed. The results give several scenarios that consist of modified suitability maps. These scenarios can be compared with the original suitability map to examine the modifications in the suitability areas classes, enabling the assessment of the number of pixels that have changed classes.

In addition, the root mean square error (RMSE) may be performed. RMSE is a very common GIS validation method that is performed in interpolation techniques evaluation (Lu and Wong, 2008, Bater and Coops, 2009) in order to determine the error propagation through spatial multi-criteria decision analysis (Malczewski, 1999; Gómez-Delgado and Barredo, 2005; Fernández and Lutz, 2010). The RMSE also allows estimating the percent of error (PE) in each scenario compared to the original suitability map, through the following equations (Lu and Wong, 2008):

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(S_i - S_{it})^2} \tag{7.1}$$

$$PE(\%) = \frac{RMSE}{(1/N)\sum_{i=1}^{N} S_{it}} \times 100 \tag{7.2}$$

Where RMSE is the mean of the squared difference between the true value (S_{it}, the suitability of the original map) and the measured value (S_i, the suitability of the scenario map); N is the number of evaluated pixels and PE (%) is the proportion of RMSE of each scenario compared to the average of original suitability. RMSE and PE represent an improvement in results analysis, to evaluate the error apportioned by changed criteria weights.

3 Case-study

3.1 Evaluation Phase

The described SMCA method was applied to a case-study in Catalonia (NE of Spain). The region is characterized by a diverse morphology, being mostly mountainous on the north (Pyrenees) and flat at the centre and the coast (Figure 7.2). The region is also characterized by the presence of the littoral mountain system, between the central

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depression and the coast. Also, the Ebro River catchment and its delta are important systems in the south of Catalonia.

The agricultural areas are mainly situated in the central depression, in the Ebro River catchment and in some coastal areas, while the population is concentrated in the coastal areas.

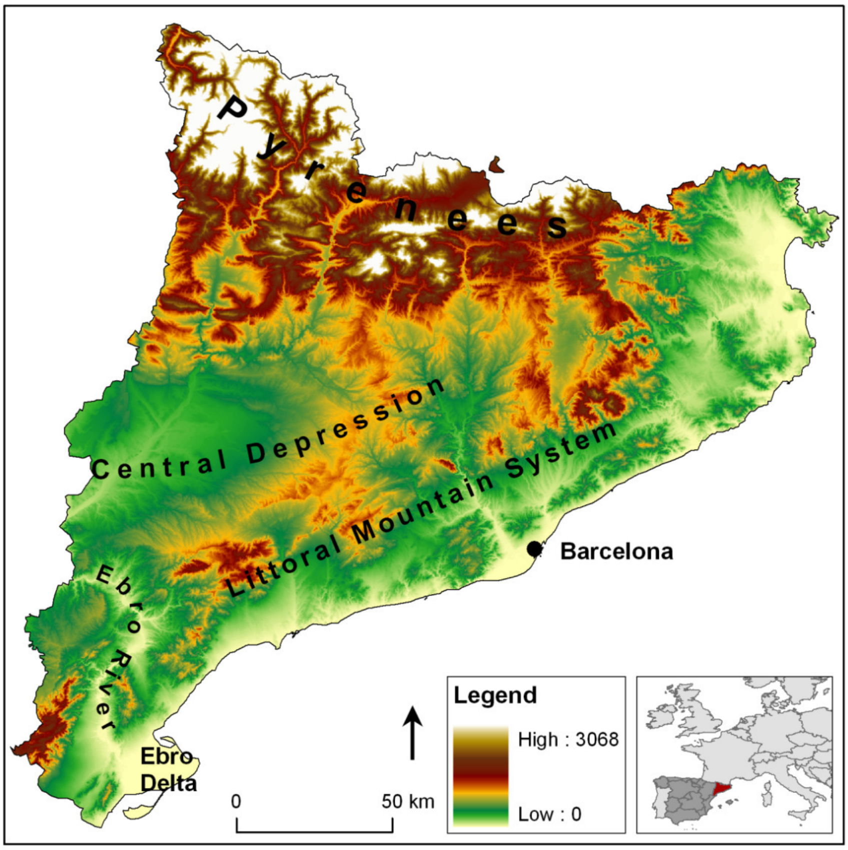

Figure 7.2. Location map of Catalonia.

The definition of the most suitable agricultural sites for sewage sludge application is a complex issue that involves considering a large set of aspects or criteria. Criteria selection was based on literature data, legislation and experts' opinion. The latter refers to the estimation of potential impacts or alterations in the environmental matrices based on an understanding of the properties of the pollutants in sewage

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sludge, soil characteristics and landscape parameters. Considering these factors, two groups of criteria were defined: human exposure and environmental (Table 7.1).

Table 7.1. Criteria tree for suitability analysis

Criterion	Sub criterion	Maps
Environmental (Contamination)	Soil	Texture, pH, Carbonates, Organic Matter, Metals
	Surface Water	Distance to Waterbodies, Slope, Temperature, Rainfall
	Groundwater	GW Vulnerability
Exposure (Human health)	Direct	Distance to Urban Areas
	Indirect	Crop Type

The exposure criterion was mainly related to human exposure to pollutants and their likelihood of producing damage to human health. The exposure criterion had two subcriteria:

- Distance to urban areas, as sludge may generate important bad odours.
- Crop types, as they may accumulate differently pollutants.

The environmental criterion expresses the likelihood of contaminating soils, surface water and groundwater, when soils are amended with SS. The following soil and landscape parameters were considered:

- Texture: Soil texture was classified according to the European Soil Database (European Communities, 2006). In this regard, a soil with high clay content was considered to have a better structure than a sandy soil.
- Organic matter (OM) content: It is well known that organic matter provides a better soil structure. In addition, organic matter degradation increases the soil content in compounds of agricultural value (such as N), which are slowlier

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released than in the case of mineral fertilizers, making them available for a longer period to crop.

- pH: It is the most important factor in metals mobility and bioavailability in soil.
- Carbonates content: The presence of carbonates decreases metals mobility in soil.
- Metal concentration: These elements are naturally present in different concentrations in background soils, although they may be also originated from anthropogenic sources such as fertilizers, animal manure, sludge application, or atmospheric deposition. Soils with lower metals content were preferred for SS amendment.
- Temperature: The mean annual temperature was considered. Zones with higher temperatures have a higher rate of organic contaminants degradation, reducing also the potential of contaminating water bodies.
- Precipitation: Mean annual rainfall values were considered, as precipitation is one of the most important factors of contaminants movement through surface runoff.
- Slope: Higher slope values lead to higher rates of surface runoff.
- Hydrology: Fields far from water bodies were considered more suitable for sewage sludge amendment.
- Groundwater: The vulnerability map elaborated by the local environmental agency (ACA, 2005), in accordance with the Nitrate Directive (EC, 2010), was employed in this study.

It must be noted that, to simplify the model, the economic criterion is not presented in this case-study. The values of the economic criterion are related to the spatial position and the sewage sludge production of each WWTP. As a consequence, an economic

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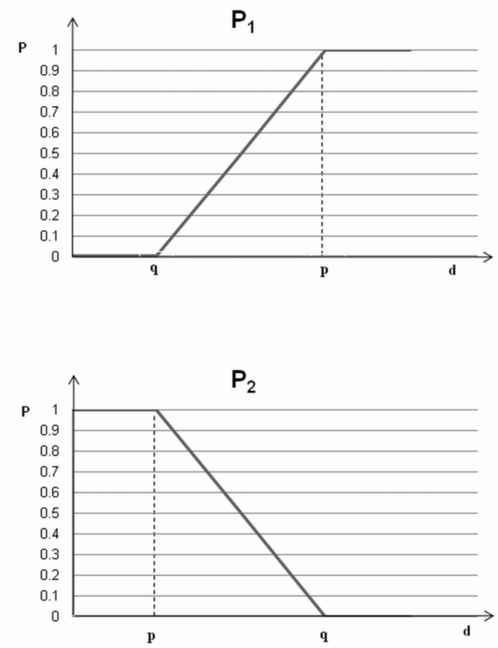
evaluation should be performed for each WWTP. In a future step, the results of this case-study should be combined with the sewage sludge production of each WWTP to perform a global evaluation.

3.2 Elaboration Phase

Based on the defined criteria, several maps were selected (Table 7.1). All the selected maps were provided by the administration. The utility of each map was defined and the utility maps were elaborated. Among the selected maps, three have discrete attributes: Crop Type, Soil Texture and GW Vulnerability. To characterize the utility of maps with continuous attributes, two types of function were employed (Figure 7.3). The preference value (*P*) is a function of the input value (*d*). For each criterion, *q* is a threshold of indifference and *p* is a threshold of strict preference (Brans and Mareschal, 2005). In those cases where criteria are preferred to be maximized (e.g., in the organic matter map), the top (P_1) preference function was applied. For criteria to be minimized (e.g., contaminants concentration in soil), the preference function is reversed and the bottom function of Figure 7.3 is applied (P_2).



$$P(d)=\begin{cases} 0 & d \le q \\ \frac{d-q}{p-q} & q < d \le p \\ 1 & d > p \end{cases}$$

$$P(d)=\begin{cases} 1 & d \le p \\ \frac{d-q}{p-q} & p < d \le q \\ 0 & d > q \end{cases}$$

Figure 7.3. Types of generalized criteria. Adapted from Brans and Mareschal (2005).

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The utility values presented in Table 7.2 were defined based on experts' opinion. Table 7.3 presents the values for the maps that discrete attributes apply.

Table 7.2. Utility values for continuous attributes

Map	Unit	Function Type	*q*	*p*
Distance to urban areas	km	P_1	0	2
Rainfall	mm yr^{-1}	P_2	1000	400
Mean Temperature (Year)	ºC	P_1	5	17
Organic matter	% dw	P_1	1	6
pH	UpH	P_1	5	8
Carbonates	$\%CaCO_3$	P_1	5	20
Metals concentration in soil	mg kg^{-1}	P_2	M*	0
Slope	%	P_2	15	8
Distance to waterbodies	m	P_1	0	500

* M is the maximum level defined by the legislation (MAPA, 1990), and it is dependant on each specific element.

Table 7.3. Utility values for discrete attributes

Crop type	Preference	GW Vulnerab.	Preference	Soil Texture	Preference
Cereal	1.00	No aquifer	1.00	Very Fine	1.00
Fruit	0.90	Low	0.90	Fine	0.80
Cabbage	0.60	Medium	0.60	Medium Fine	0.60
Pasture	0.30	High	0.20	Medium	0.40
				Coarse	0.20

Each of the cited criteria was defined as a raster map of standardized values between 0 and 1, where higher values represent more suitable areas. The model was integrated in IDRISI (Clark Labs, 2001) and the raster grid definition was 200 x 200 m. This scale was found to be adequate for the studied region. The total number of evaluated pixels was of approximately 260.000. The normalized input maps are shown in Figure 7.4. All the input factors were assumed to be independent of each other.

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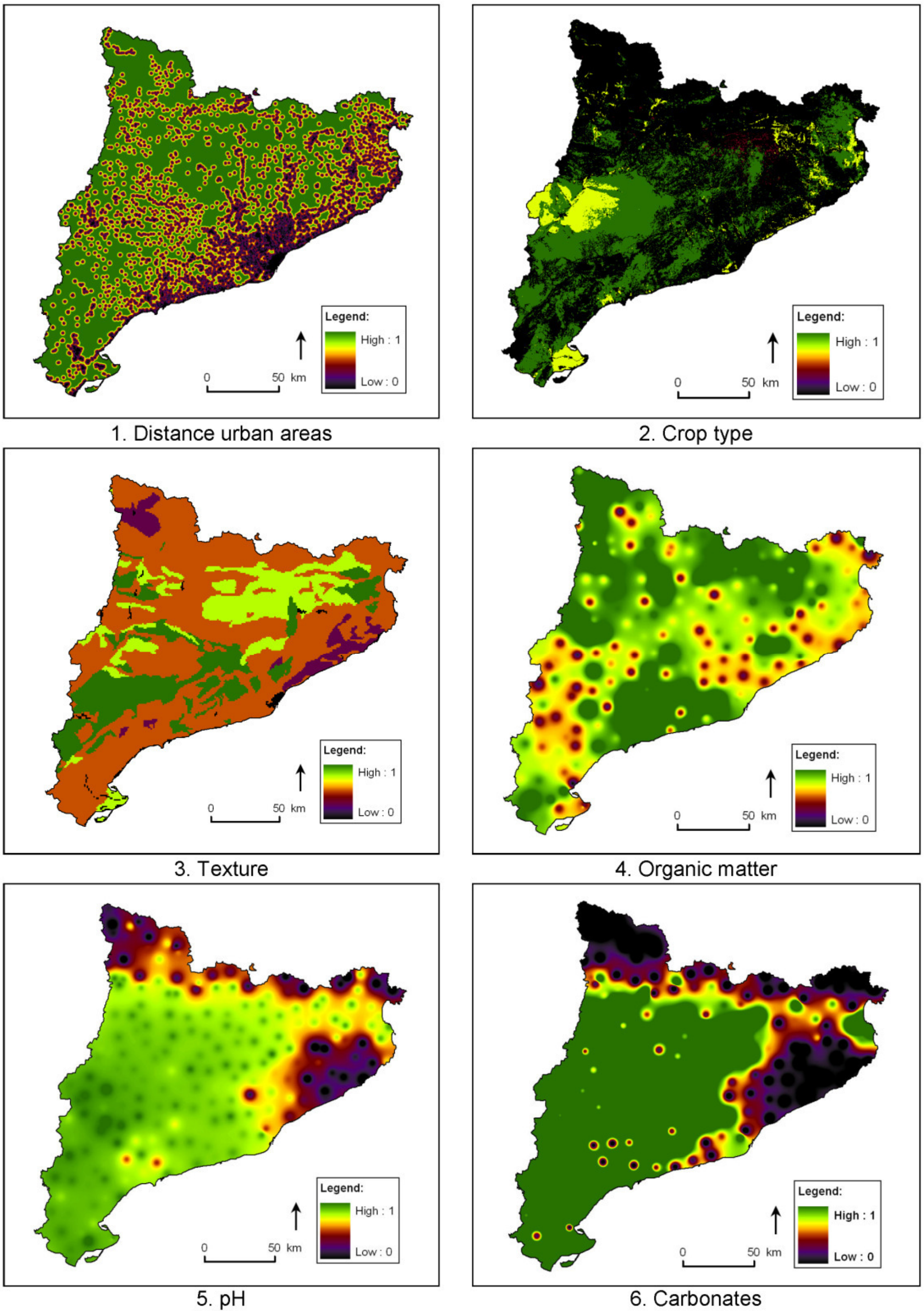

Figure 7.4. Normalized input maps of the evaluation (continues).

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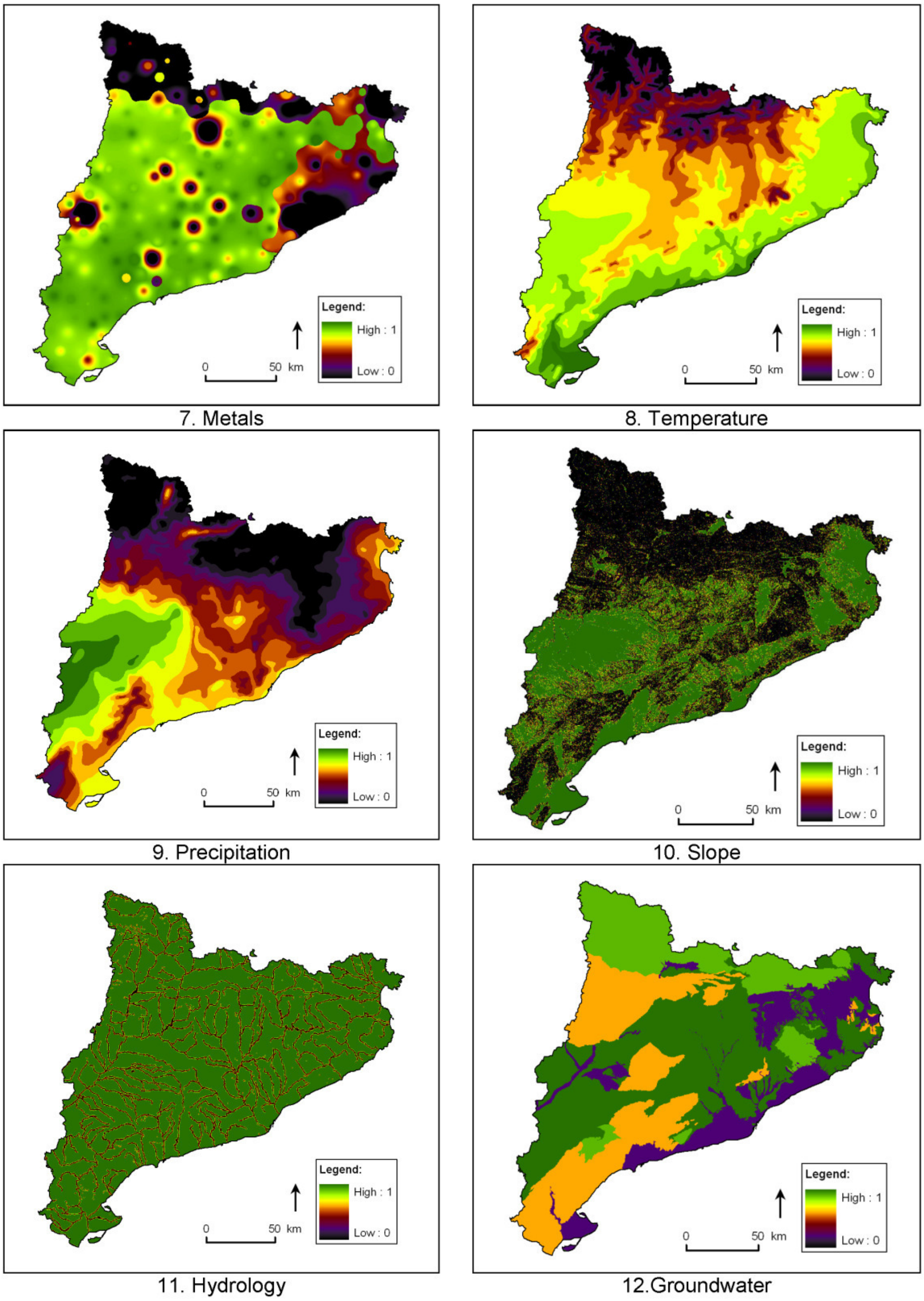

Figure 7.4. Normalized input maps of the evaluation (continued).

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3.1 Integration

As previously described, there are several methods for implementing a SMCA. The LSP method was selected because of the easiness for treating the available data and the possibility of considering simultaneity and replaceability. The use of AHP was rejected due to the large amount of criteria and the complexity of the case-study.

LSP method is based on mathematical models that use Generalized Conjunction/Disjunction (GCD) and other continuous preference logic functions (Dujmovic, 2007), taking into consideration the different levels in the hierarchy of criteria, weights and constraints, over those criteria. Three LSP aggregators were selected for the present case-study: partial disjunction (DA), arithmetic mean (A) and partial conjunction (CA).

The DA aggregator translates the replaceability of the criteria (partial disjunction), and a high value in only one input leads to a high output value. For instance, a good soil structure is translated by a high organic matter value or by an appropriate texture (complementarity). A soil with a poor texture (sandy, sandy loam soil), that has been amended for several years, may have a high OM content and, in consequence, a good structure. In contrast, the CA aggregator represents a partial conjunction between the factors. This means that both inputs must be high to result in a high output (simultaneity). For example, a good score in “slope” and “hydrology” is needed to get a good score in the “relief” map. In other words, to avoid surface water contamination a low terrain slope is needed but also the distance to waterbodies must be sufficiently high. For the A aggregator, weighted arithmetic mean is applied. The model rules (aggregators and weights) are shown in the Figure 7.5.

Most of of the employed aggregators were “CA”, representing a partial conjunction between the factors (Figure 7.5). This means that both inputs must be high to result in a high output (simultaneity).

The model was integrated based on the previously described steps, considering only the areas with agricultural land use of Catalonia.

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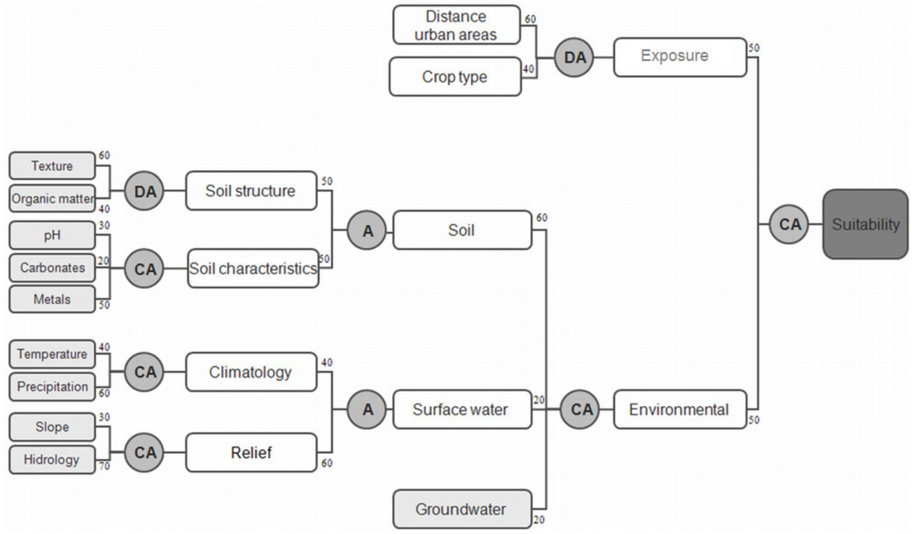

Figure 7.5. Schematic representation of the model implementation. The values (numbers) indicate the weights of each map (in %) and the letters in the circles the aggregator: partial disjunction (DA), arithmetic mean (A), partial conjunction (CA).

4 Results and discussion

The integrated model, based on the previously described methodology, was applied to the Catalan agricultural areas. For this evaluation, four suitability classes were defined (Figure 7.6). The areas with values above 0.7 (medium to very high scale) were considered suitable for receiving sewage sludge. It should be noted that these areas represent more than 680,000 hectares, being sufficient for managing the amount of sludge produced in Catalonia. The suitable areas are mainly located in the central depression (center-west) and in the Ebro River catchment (south). The central depression was found to be the best area for sewage sludge amendment in Catalonia. In fact, this region represents the most important agricultural area of Catalonia nowadays.

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4.1 Global Sensitivity Analysis

To evaluate the importance of the input maps, a global sensitivity analysis (GSA) (Campolongo et al., 2000) was performed. The analysis took into account the variability of the results in the whole studied region, based on the maps histograms. According to Campolongo et al. (2000), standardized regression coefficients (β) are an alternative to the variance-based methods that are only effective for linear or quasi-linear models (coefficient of correlation $R^2 \geq 0.70$). The β values were calculated with the software Analytica (Lumina, 2008).

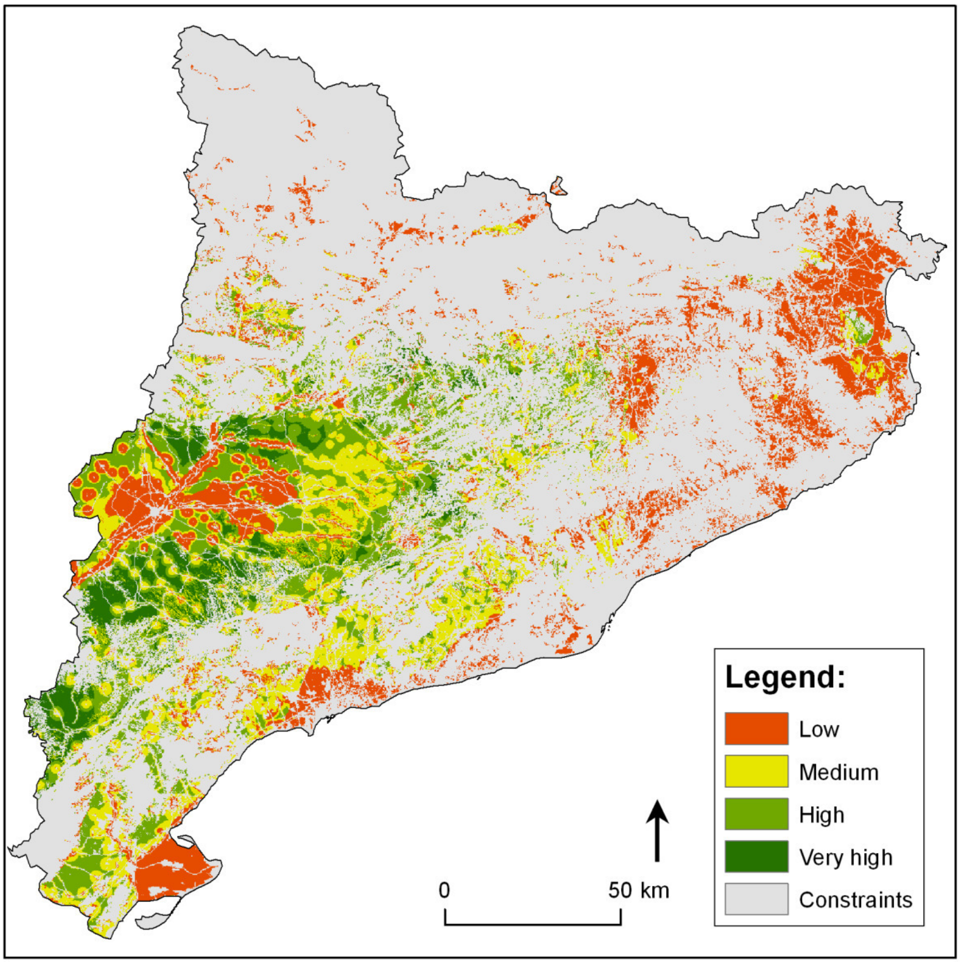

Figure 7.6. Spatial representation of the suitability of Catalan agricultural areas. Suitability classes: low (< 0.7), medium (0.7-0.8), high (0.8-0.9), very high (0.9-1.0).

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Monte Carlo sampling was performed for 5,000 iterations and high correlation values (R^2=0.80) were obtained, indicating that the model acts linearly and higher order effects are not important. SA indexes are shown in Table 7.4.

Table 7.4. Global sensitivity analysis results.

Map	β value	S value
Groundwater	0.48	23%
Distance urban areas	0.43	19%
Metals	0.33	11%
Crop type	0.30	9%
Slope	0.28	8%

The Groundwater map explained 23% of the variability of the results, while the map Distance to urban areas explained 19% of results variability (Table 7.4). Other maps of pronounced importance were Metals, Crop type and Slope. Most of the variability of the results was explained by 5 input maps, being the other maps of relative low importance (<8%). Due to this fact, for the application of the exploratory method, only the 5 maps with the highest importance were considered.

4.2 Exploratory method

The exploratory method was performed to evaluate the variation in a suitability map results when different changes are introduced. For that, 5 scenarios were created. These scenarios corresponded to the elimination of each of the most important input maps in model evaluation, and the redistribution of weights. One scenario was created for each of the most sensitive maps defined in the previous step (Table 7.4). The resulting suitability maps were reclassified into the four previously defined suitability classes (defined in Figure 7.5). Then, the maps were compared to the original suitability maps. Figure 7.6 shows the differences in suitability classes for each scenario. The pixels with increased classes presented a better score in the scenario map than in the original map (suitability). In contrast, the pixels with decreased classes had a lower score in the scenario map than in the original map.

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Of the five evaluated scenarios, Scenarios 1, 2 and 3 presented the highest number of pixels with changed classes. This may be due to the fact that these three scenarios are related to the maps with a high hierarchy in the model implementation (Figure 7.4).

Scenario 1 (without Groundwater map) presented a similar quantity of pixels that increased and decreased their values, probably because vulnerable areas had increased while non-vulnerable areas had decreased suitability values. Scenario 2 (without Distance urban areas map) presented the higher number of pixels with increased suitability, while Scenario 4 (without Crop type map) had the higher number of pixels with decreased values. These two last scenarios corresponded to two input maps that are connected in the model structure and had a disjunctive aggregator that establish a compensative relationship (Figure 7.4). As the map Distance to urban areas values are lower than the Crop type map, the extraction of the Distance to urban areas map leaded to higher social scores and vice versa.

Scenarios 3 (without Metals map) and 5 (without Slope map) presented the lower variation on suitability results, and in general had increased values when compared to the original evaluation. This fact is a consequence of the application of a conjunctive aggregator that tends to obtain the lower score between the aggregated maps. The increased pixels corresponded to low values for the eliminated map.

To enhance the spatial evaluation, the root mean square error (RMSE) was calculated. The application of the RMSE method allowed the evaluation of the exact error apportioned due to the elimination of each map.

The results of RMSE and percent errors are shown in Table 7.5. Scenario 4 (without Crop type map) presented the highest error percentage. This means that the elimination of the Crop type map would lead to the higher error in the output. In fact, this agrees with the results presented in Figure 7.6 that show a pronounced decrease in the output values when this map was not considered. The other scenarios were responsible for between 6 and 10% of the final map error. The results of this evaluation will be useful for model improvement and model validation in a more representative area.

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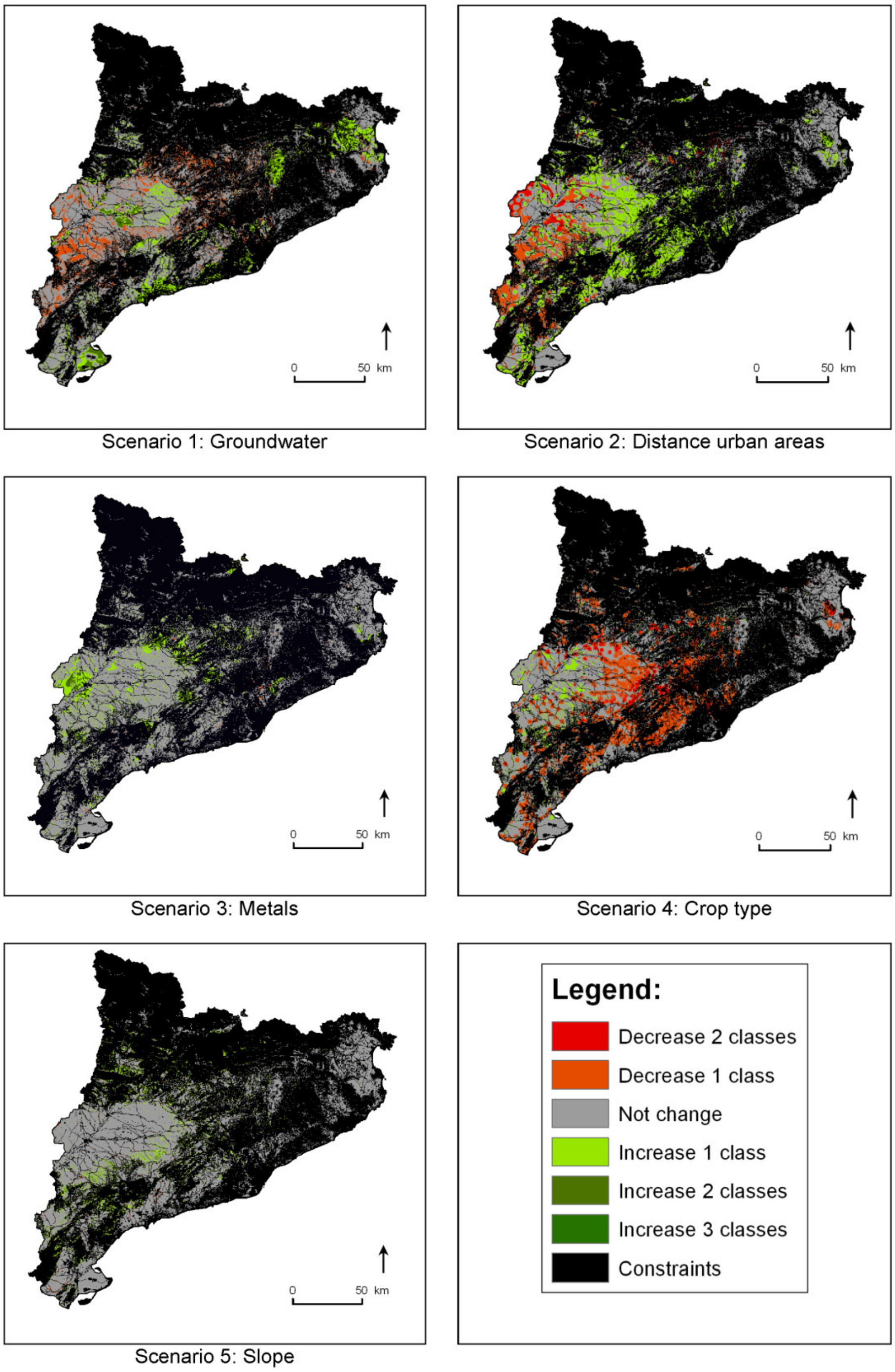

Figure 7.6. Spatial representation of the differences between pixels classes for each scenario. Each map caption indicates the maps extracted from the evaluation.

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Table 7.5. Root Mean Square Error and percent error results

Scenario	Eliminated map	RMSE	Percent Error
SA1	Groundwater	0.055	7.5%
SA2	Distance urban areas	0.071	9.7%
SA3	Metals	0.045	6.1%
SA4	Crop type	0.182	24.9%
SA5	Slope	0.063	8.7%

5 Conclusions

In this chapter, a Spatial Multicriteria Model was developed to support decisions regarding the application of sewage sludge on agricultural soils. The case-study results point out that the suitable areas are sufficient for the management of this residue.

Different tools for results evaluation were applied. These tools showed different trends for the two performed evaluations. This fact was expected as GSA evaluates the histograms of the whole region, while the exploratory method assesses the variations in each pixel.

Groundwater map was found to be responsible for most of the variability in the results (GSA). Groundwater contamination is mainly related with the points that showed low and very low scores in the central depression. These points correspond to zones above aquifers that are highly vulnerable to nitrate contamination. GW contamination map also influenced the low scores obtained close to the Ebro River Delta. However, the error apportioned by the elimination of this map was of 7.5%. This is explained by the fact that this map acted as a restriction in the vulnerable areas. These areas were not representative of the total agricultural areas and, in consequence, a low error was apportioned by the elimination of this map.

Crop type was found to be another important factor of the evaluation. Indeed, this result was expected as in the evaluation of the suitability of agricultural fields the crop type should be one of the most important factors of the evaluation. Most of the suitable

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areas corresponded to fruit and cereal fields, while cabbage and cattle fields corresponded to areas with low and very low suitability values. The elimination of this map represented the higher RMSE. However, this map is responsible for only 9% of the variability in the results. This may be explained by the fact that most of the agricultural areas correspond to fruit and cereal fields, which were assumed to be the most suitable crop types in model development. Because of that, the variability of this map in the region was low, leading to low β values, but the values apportioned by this map were high (Figure 8.3), leading to a high RMSE.

It has been demonstrated that the application of tools for results evaluation enhance the models understanding in environmental decision support systems. These tools may be applied to evaluate different spatial models and give an improved understanding of the frameworks. In addition, the results obtained may be useful for the elaboration and the development of future studies in a regional scale, considering a more accurate map resolution and model simplification, when necessary. Also, a simplified version of the model could be applied for integrating economic and environmental costs to manage a defined sewage sludge.

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Chapter 8

Application of Bayesian networks to land suitability classification

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1 Introduction

In recent years total production of sewage sludge (SS) in many countries has increased due to improved rates of sewage treatment and demographic changes. Managing an increased production of sewage sludge (SS) has become an issue of great interest. As for any residue, the reuse or recycling of SS is preferred to its disposal. Thus, amending soils with SS is a preferred route in several European countries, due to its fertility benefits to soil and increased crop production. SS has an appropriate content of composts of agronomic value, such as nitrogen, phosphorous and organic matter. The use of treated SS as an organic fertilizer improves soil structure and acts as a buffer to soil contamination due to slower release of nutrient compounds from the stabilized sludge than from mineral fertilizers (Passuello et al., 2011).

However, the amendment of soils with SS has some associated risks, due to the presence of contaminants in the sludge matrix, such as heavy metals (Aubain et al., 2001; Metcalf and Eddy, 2003) and Persistent Organic Pollutants (POPs) (Eljarrat et al., 2003; Harrison et al., 2006). These contaminants may be transferred from soil to crops and enter the human food chain. While farmers' interests are the increase in productivity and the profits obtained from the practice, authorities must assure that amendment is performed in proper health and environmental safety and with low impact to the community. Also, the evaluation of the availability of areas to apply sewage sludge is essential because while SS production increases, the availability of land is limited.

To define the suitability of the agricultural areas is a complex task that involves several issues. Soil suitability should have a qualitative classification, instead of being represented by a single number. However, soil parameters are measured numerically, through concentrations for instance. Also, some parameters, such as crop type, are discrete and no value applies to those cases. The integration of the parameters of interest is a complicated task due to the fact that these parameters are incomparable and sometimes incommensurate.

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In addition, the lack of knowledge on the representation of natural systems is of concern and a high uncertainty is associated to model development. In this case, the modeller must define which processes and objectives consider, facilitating interdisciplinary communication and applying different sources of knowledge. Bayesian networks (BNs) can explicitly deal with uncertainty (van Kouwen et al., 2008). This method is able to integrate knowledge from different disciplines, aggregating system complexity to the level that is appropriate, representing and communicating uncertainties.

BNs have been successfully applied on land management and land use policy. Cain et al. (2003) investigated whether Bayesian networks, together with approaches to help people use them, could provide the generic framework to develop a DSS for agricultural system management. BNs were perceived to help the planning process as they allowed policy makers to develop complex systems from a multi-disciplinary perspective. Ticehurst et al. (2011) compared BNs with conventional statistical analysis to explore the usefulness of BNs for the analysis of social data sets in a case-study of natural resources management. The authors found that BNs results were more easily interpreted and communicated then traditional statistical outputs. Also, Bacon et al. (2002) performed a two stage model of land use change. BNs were applied to explore the relations between personal satisfaction and the costs for the landholders to change their land use. The authors argued that BNs are a powerful and robust method to bridge the gap between stakeholders and experts.

BNs are able to capture the described factors through influence diagrams that describe causal relationships between input parameters and indicators of risk levels. The probability theory underlying BNs provides a consistent calculus for uncertainty inference, and so a BN-based integrated model can be a useful tool to analyse the implications of uncertainties for future decisions (Lerner et al., in press). Due to all the described reasons, BNs was selected as a tool for developing a soil classification model.

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There is no application of BNs for land suitability classification found in the scientific literature. This analysis, coupled with GIS, gives insights for policy makers and farmers on the best management options for the studied area. In this chapter, the application of BN in land suitability classification, in order to define the best agricultural areas to amend with sewage sludge is described. A case-study of sewage sludge application in agricultural land in North-Eastern part of Spain (Catalonia) has been used to describe model's application.

2 Materials and Methods

2.1 Method description

A Bayesian network is a decision analysis framework, based on Bayesian probability theory, which allows the integration of scientific and experiential knowledge, and the uncertainty associated with this knowledge (Castelletti and Soncini-Sessa, 2007). The approach involves describing a system in terms of variables and linkages, or relationships between variables, at a level appropriate to the decision making. This is achieved through representing linkages as conditional probability tables and propagating probabilities through the network to give the likelihood of variable outcomes (Murphy, 2001). Therefore, the approach ensures that the treatment of risks and uncertainties is an intrinsic part of the decision-making processes (Borsuk et al., 2004). The Bayesian network is flexible and interactive, and hence if a previously developed network does not fit a user conceptual understanding of the system, it can be adapted quickly and simply to the cognitive understanding of the user. Mathematically speaking, Bayesian networks are the Directed Acyclic Graph (DAG) where nodes represent variables, arcs between nodes represent probabilistic dependencies. A graph is called directed if the graph links have directions. A directed graph is acyclic if the graph contains no directed cycles. Bayesian Probability Networks provide a representation of multivariate probability distribution over a set of variables in an uncertain domain. Each node in the directed acyclic graph represents a random variable. These variables are conditionally dependent only on their parent nodes. Each node has a probability distribution associated with it, which represents probabilities

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given its parents states. A conditional probability table (CPT) is commonly used to store this information for each variable node.

2.2 Case-study

Catalonia (NE of Spain) is characterized by a diverse morphology, being mostly mountainous in the north (Pyrenees) and flat at the centre and the coast (Figure 8.1). Catalan agricultural areas are mainly situated in the central depression, in the Ebro river catchment and in some coastal areas, while the population is concentrated in the coastal areas. Catalonia has a population of more than 7.5 million habitants and an annual SS production of 140,000 tonnes dw. Between those, approximately 115,000 tonnes are disposed in agricultural fields. Catalonia agricultural areas sum more than 1 million hectares. More than 85% of this area is covered by fruit and cereal fields (ACA, 2008; IDESCAT, 2010).

Fig 8.1. Location map of Catalonia

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In this region, SS management in agricultural land is performed based on the best possible economic profit and legislation purposes. The local legislation regarding SS amendment on agricultural soils defines maximum levels of metals (Cd, Cr, Cu, Hg, Ni, Pb, Zn) in SS and soils, with the objective of assuring that safe levels of these contaminants are maintained into food (MAPA, 1990). In addition, areas that are vulnerable to groundwater contamination as a consequence of the pollution caused by nitrates from agricultural sources are defined for Catalonia, as established by the European Directive 91/676/CEE (CEC, 1991b). The economic costs of applying SS to land depend on the location of the field and the treatment plant. To simplify the model, the economic criterion was not assessed in this study, as an economic evaluation should be performed for each WWTP. In the future, the results of this case-study shall be combined with data from each WWTP to perform a global evaluation.

One important problem of the management nowadays is public opposition, as the population is concerned about surface water and groundwater contamination, soil and food quality and odours (Goven and Langer, 2009). Several of these factors are related to local characteristics of the land. It has been proved that the fate of organic compounds in plants depends on crop type (cereal, fruit…) and vegetal part (root, leave…) (Legind and Trapp, 2009). For example, pollutants transference to seeds in cereal fields is much lower than in the case of cabbage and pasture fields. Also, the contaminants in pasture fields are transferred to meat and milk, being biomagnified. The likelihood of contaminating waters is linked to the characteristics of the terrain, as slope, and rainfall. In consequence, some agricultural fields may be more suitable for receiving SS than others. As the production of SS is increasing and the availability of land is limited, the use of land classification tools is essential to assure that this practice is performed in safe levels during the following years.

2.3 Model development

The BN model was developed to classify the suitability of different agricultural areas to amend with sewage sludge. The model was intended to represent causal relationships

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between the terrain characteristics and the identified problems related to the amending practice.

Several workshops were organized with stakeholders and environmental experts with the objective of identifying the main problems related to practice, investigating the related impacts according to different soil and landscape characteristics. The main problem identified by the stakeholders is the accomplishment of the legislation, especially regarding the areas affected with groundwater contamination. The possibility of contaminating the human food chain, due to the contamination of crops and water, was pointed by some stakeholders. In contrast, an experts group gave a broader and deeper evaluation of the problem. This group was formed by chemists, ecologists, environmental scientists, engineers, geographers and experts in soil science. The group found two main criteria for this problem: an exposure criterion, related to the direct human exposure of the contaminants present in sewage sludge, and an environmental criterion, related to the protection of soil and water bodies. The defined group of criteria considers both the problems identified by stakeholders and experts, assessing all the soil and landscape characteristics that are relevant to protect the environment, accomplishing the levels established by the local, national and European legislation. The criteria are described in the Tables 8.1 and 8.2.

In a previous study, Passuello et al. (2011) evaluated the same problem through the application of the LSP method in GIS. LSP method is based on mathematical models that use Generalized Conjunction/Disjunction (GCD) and other continuous preference logic functions (Dujmovic, 2007), taking into consideration the different levels in the hierarchy of criteria, weights and constraints over those criteria. This method allows considering if criteria are complementary or replaceable. However, this method does not allow assessing the uncertainty related to any decision process.

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Table 8.1. Description of model input parameters

Input	Motivation	Source	States
Distance to urban areas	Considered an important factor for reducing bad odours.	Defined by experts	<1.4km, 1.4-1.6 km, 1.6 - 1.8 km, > 1.8km
Crop type	Different crop types have different potential of accumulating contaminants.	Legind and Trapp (2009)	Cereal, Fruit, Cabbage, Pasture
Texture	Soil texture was classified according to the European Soil Database (European Communities, 2006). In this regard, a soil with high clay content was considered to have a better structure than a sandy soil.	European Communities (2006)	Very fine, Fine, Medium fine, Medium, Coarse
Organic matter (OM) content	Organic matter provides a better soil structure. OM is related to soil organic carbon by the Van Bemmelen factor.	Jones et al. (2004)	<4.5%, 4.5-5.0%, 5.0-5.5%, >5.5%
pH	Between soil parameters, pH is the most important factor in metals mobility and bioavailability in soil.	Porta Casanellas et al. (2003)	<7.1, 7.1-7.4, 7.4-7.7, >7.7
Carbonates	Carbonates decrease metals mobility in soil.	Porta Casanellas et al. (2003)	<15.5%, 15.5-17.0%, 17.8-18.5%, >18.5%
Metals concentration	Soils with lower metals content are preferred for SS amendment. These levels are controlled by the Spanish Royal Decree 1310	MAPA (1990)	See Table 8.2.
Temperature	Considers the mean annual temperature. Zones with higher temperatures present a higher rate of organic contaminants degradation.	MAPA (2008)	<13.4ºC, 13.4-14.6ºC, 14.6-15.8ºC, >15.8ºC
Precipitation	Regards the mean annual precipitation values. Higher precipitation rates are responsible for contaminants movement through lixiviate.	MAPA (2008)	<460mm yr^{-1}, 460-520mm yr^{-1}, 520-580 mm yr^{-1} , >580 mm yr^{-1}
Slope	Higher slope values lead to higher rates of surface runoff. The classification of the European Digital Archive on Soil Maps of the World was considered.	EuDASM (2008)	<8.7%, 8.7-9.4%, 9.4-10.1%, >10.1%
Hydrology (distance to river bodies)	Fields far from water bodies are considered more suitable for sewage sludge amendment.	Metcalf & Eddy (2003)	<350m, 350-400m, 400-450m, >450m
Groundwater	In accordance with the Nitrate Directive (EC, 2010), the vulnerability map elaborated by the local environmental agency was employed in this study.	ACA (2005)	No vulnerable, Low, Medium, High

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In order to better represent knowledge uncertainty, BNs were selected to evaluate this problem. While LSP calculation gives an exact value for each pixel, BNs considers the uncertainty of the system parameters. In consequence, BNs results allows more confident decisions in the sense that it captures the uncertainty that is present in the decision model, such as when trying to translate classes into values. This is an interesting point as many stakeholders prefer working with classified data such as soil type or nutrient levels, etc.

Table 8.2. Description of the states for metals concentration in soil (mg kg^{-1} dw)

Metal	Acid soil	Basic soil
Cadmium	<0.1, 0.1-0.2, 0.2-0.3, >0.3	<0.3, 0.3-0.6, 0.6-0.9, >0.9
Copper	<5, 5-10, 10-15, >15	<21, 21-42, 42-63, >63
Nickel	<3, 3-6, 6-9, >9	<11.2, 11.2-22.4, 22.4-33.6, >33.6
Lead	<15, 15-30, 30-45, >45	<30, 30-60, 60-90, >90
Zinc	<15, 15-30, 30-45, >45	<45, 45-90, 90-135, >135
Mercury	<0.1, 0.1-0.2, 0.2-0.3, >0.3	<0.15, 0.15-0.30, 0.30-0.45, >0.45
Chromium	<10, 10-20, 20-30, >30	<15, 15-30, 30-45, >45

The framework applied to this study is shown in Figure 8.2. First, all the input maps are classified, according to the levels defined in Tables 8.1 and 8.2. Then, the model is integrated and the probability of each suitability result is presented in maps. This is a key point of this evaluation as each pixel has a corresponding probability of reaching each of the classes.

The BN (Figure 8.3) was developed intending to maintain the relationships presented at Passuello et al. (2011) between the selected criteria.

The diagram presents the parents (causes) on the left, and the children (consequences) on the right side. This diagram should be interpreted according to these hierarchical relationships. For example, higher temperatures are responsible for higher rates of contaminants degradation, being more suitable than lower temperatures. In addition, surface runoff rates are lower when lower precipitation

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levels are observed. These two factors combined with other factors (slope, hydrology) may prevent rivers contamination in the studied area.

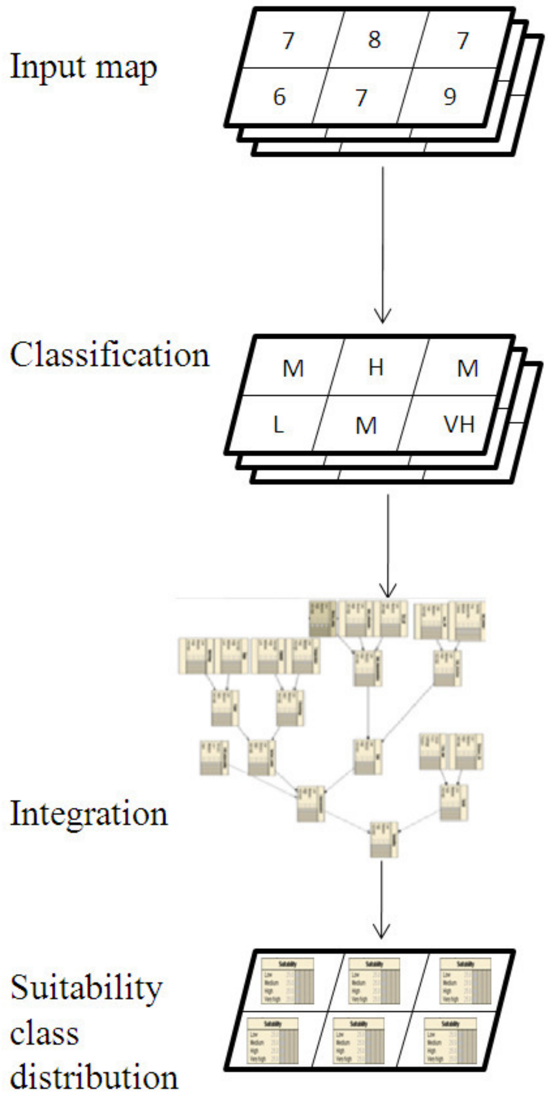

Figure 8.2. Framework for suitability classification for the BN model.

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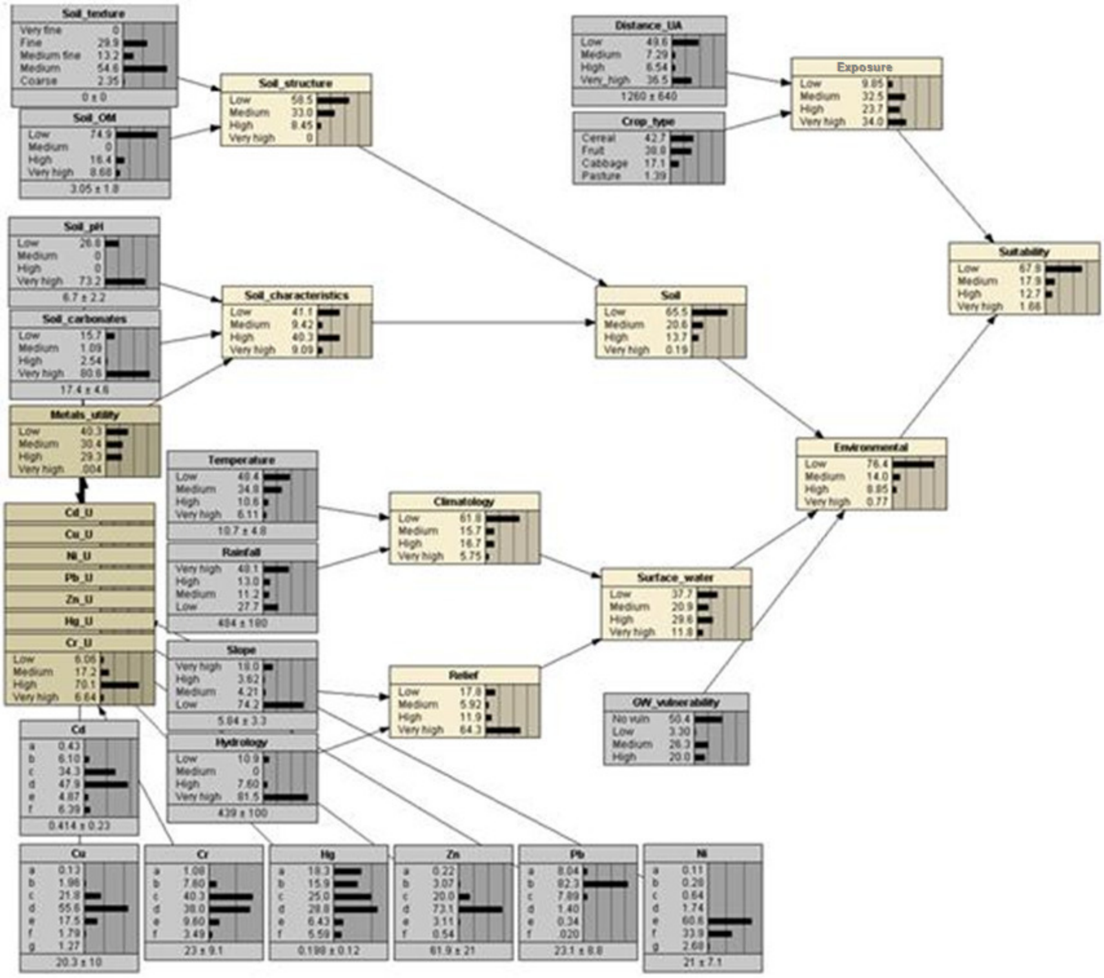

Figure 8.3. Land classification BN showing the structure and states. The BN was populated with data from the study area.

To translate the LSP rules into CPTs, three steps were followed:

(i) Classify each input value according to the previously defined ranges;

(ii) Run the LSP method for all the combinations of these ranges;

(iii) Calculate the outputs probability which is used to populate CPTs. One example of a CPT is given in Table 8.3.

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Table 8.3. CPT for the node "Soil structure"

Parent classes		Child classes			
Soil_texture	**Soil_OM**	**Low**	**Medium**	**High**	**Very high**
Very fine	Low	0.00	0.00	0.87	0.13
	Medium	0.00	0.00	0.00	1.00
	High	0.00	0.00	0.00	1.00
	Very high	0.00	0.00	0.00	1.00
Fine	Low	0.00	1.00	0.00	0.00
	Medium	0.00	0.91	0.09	0.00
	High	0.00	0.00	1.00	0.00
	Very high	0.00	0.00	1.00	0.00
Medium fine	Low	1.00	0.00	0.00	0.00
	Medium	0.91	0.09	0.00	0.00
	High	0.00	1.00	0.00	0.00
	Very high	0.00	0.55	0.45	0.00
Medium	Low	1.00	0.00	0.00	0.00
	Medium	1.00	0.00	0.00	0.00
	High	0.64	0.36	0.00	0.00
	Very high	0.00	0.91	0.09	0.00
Coarse	Low	1.00	0.00	0.00	0.00
	Medium	1.00	0.00	0.00	0.00
	High	0.82	0.18	0.00	0.00
	Very high	0.00	1.00	0.00	0.00

In this case, the LSP aggregator is a DA, which means that criteria act complementary. The higher suitability classes are given for the very fine soil and the higher OM contents. For example, if the soil texture is "medium fine", there is a high probability of a low suitability if OM is "low" (1.00, or 100% of the cases) or "medium" (0.91). In contrast, for the same soil texture, a very high OM content could lead to a "medium" (0.55) or "high" (0.45) suitability. Note that the values in each line must sum 1 to represent all the possible probabilities.

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3 Results and discussion

The suitability evaluation was performed for an area of more that $2.4x10^6$ ha, generating a great amount of results. The understanding of these results is improved when we evaluate the different types of information given by the model. For that, we first present the general trends regarding soil suitability for receiving sewage sludge, which are represented in this case by the most probable (MP) class for each evaluated area (Figure 8.4).

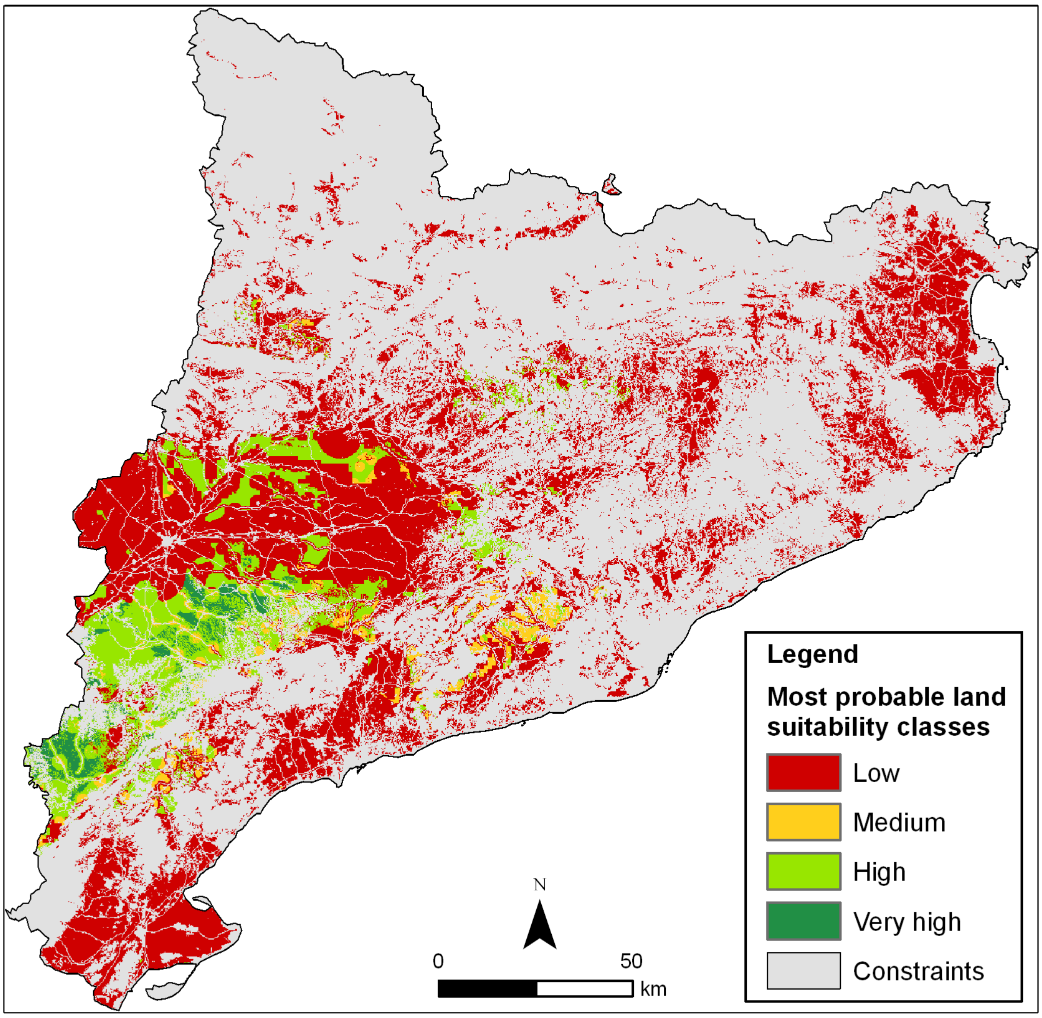

Figure 8.4. Most probable land suitability classes

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The southern and extreme northern parts of the central depression as well as the upper part of the Ebro River catchment are the most suitable ones. These areas present High and Very high MP classes, indicating that are highly suitable for receiving sewage sludge as an organic amendment.

The majority of the areas present a Low suitability as the MP value. This may be due to the fact that the model is highly restrictive, as shows Figure 8.3. However, a broader understanding of the likelihood of these areas to have each classification is needed. The probability maps illustrate the likelihood of reaching each class for each zone. They give a better understanding of the classification and the uncertainty related to each class. The results are shown in Figure 8.5.

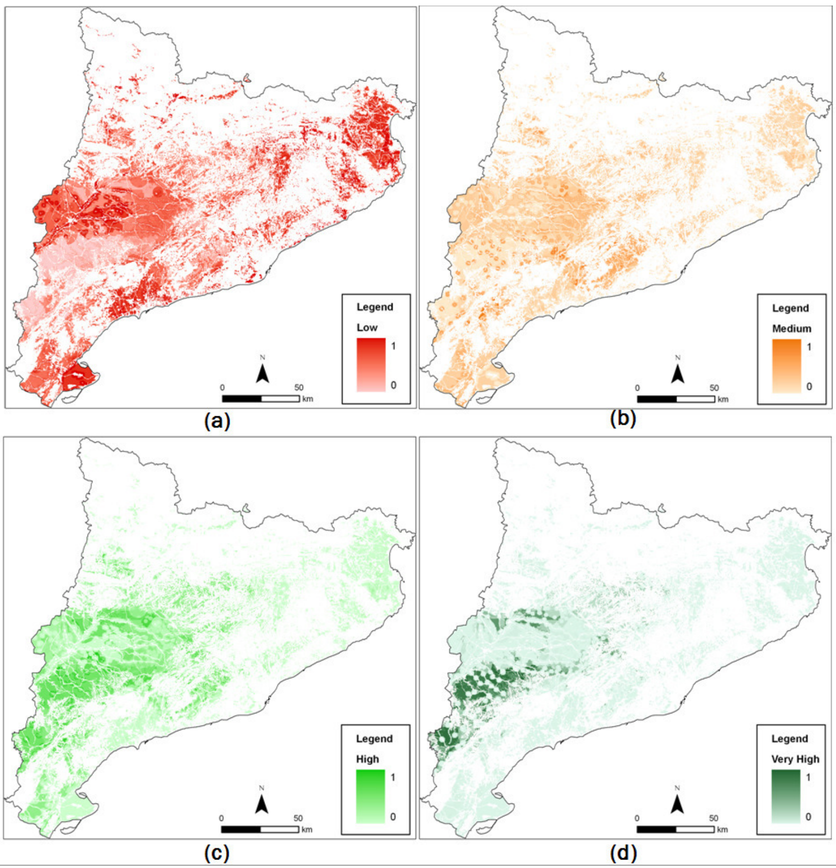

Figure 8.5. Probability maps defining the likelihood of the different suitability ranges for (a) Low, (b) Medium, (c) High and (d) Very high suitability classes.

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The probability maps (Figure 8.5) present different trends when compared to the MP map (Figure 8.4). This fact was expected, as the information in the MP map is incomplete because it does not consider the model's uncertainty.

The central depression, for instance, presents different probability values, depending on the zone. The fields in dark red on Figure 8.5a indicate a suitability lower than the adjacent regions. Figure 8.5b shows that many of the central areas could be classified as medium suitability (dark orange areas). The maps presented on Figure 8.5c and 8.5d confirm that the most suitable areas are the southern and extreme northern parts of the central depression and the upper part of the Ebro river catchment, being the first ones more suitable than the last one.

The comparison of the maps in Figure 8.5 indicates a high uncertainty related to the classification in most of the areas, especially for those with Low and Medium classes. To better evaluate the uncertainty related to land classification, the CPT of each point should be assessed. To present the CPTs of all the evaluated data is not possible due to the great amount of data evaluated and generated by the model. Instead, we selected a subset of the results that we believe should be discussed to improve results understanding. The description of each evaluated case and its geographical position are shown in Table 8.5 and Figure 8.6, respectively. The CPTs are presented in Figures 8.7 and 8.8.

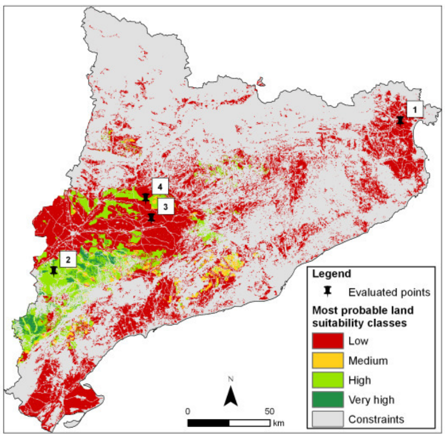

Figure 8.6. Geographical position of the 4 selected cases.

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Table 8.5. Cases description

Case number	Description
1	NE part of Catalonia, Low suitability, low uncertainty. Cereal field, acid soil, high GW vulnerability, close to cities.
2	Southern part of the central depression, High to Very high suitability, high uncertainty. Fruit field, basic soil, no vulnerable area, far from cities.
3	Central part of the central depression, Low suitability, medium uncertainty. Cereal field, basic soil, area with medium GW vulnerability, close to cities.
4	Northern part of the central depression, High suitability, medium uncertainty. Cereal field, basic soil, no vulnerability, close to cities.

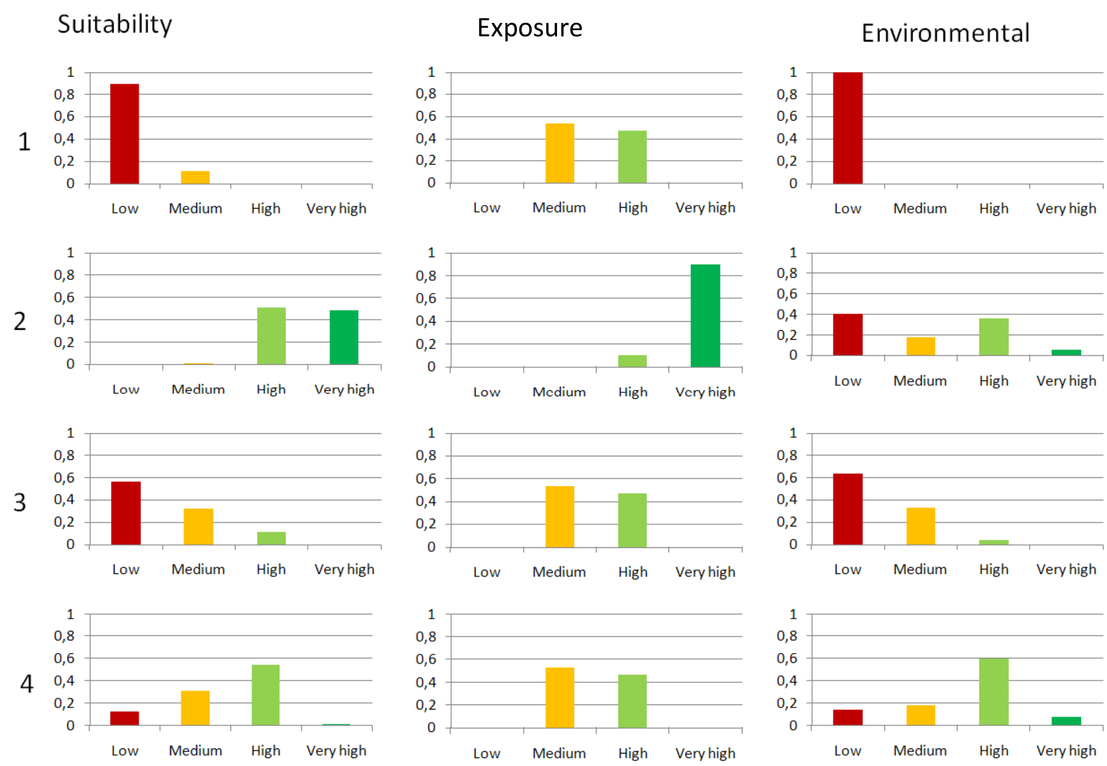

Figure 8.7. CPTs, represented as distributions, for the nodes Suitability, Social and Environmental, for the 4 selected cases.

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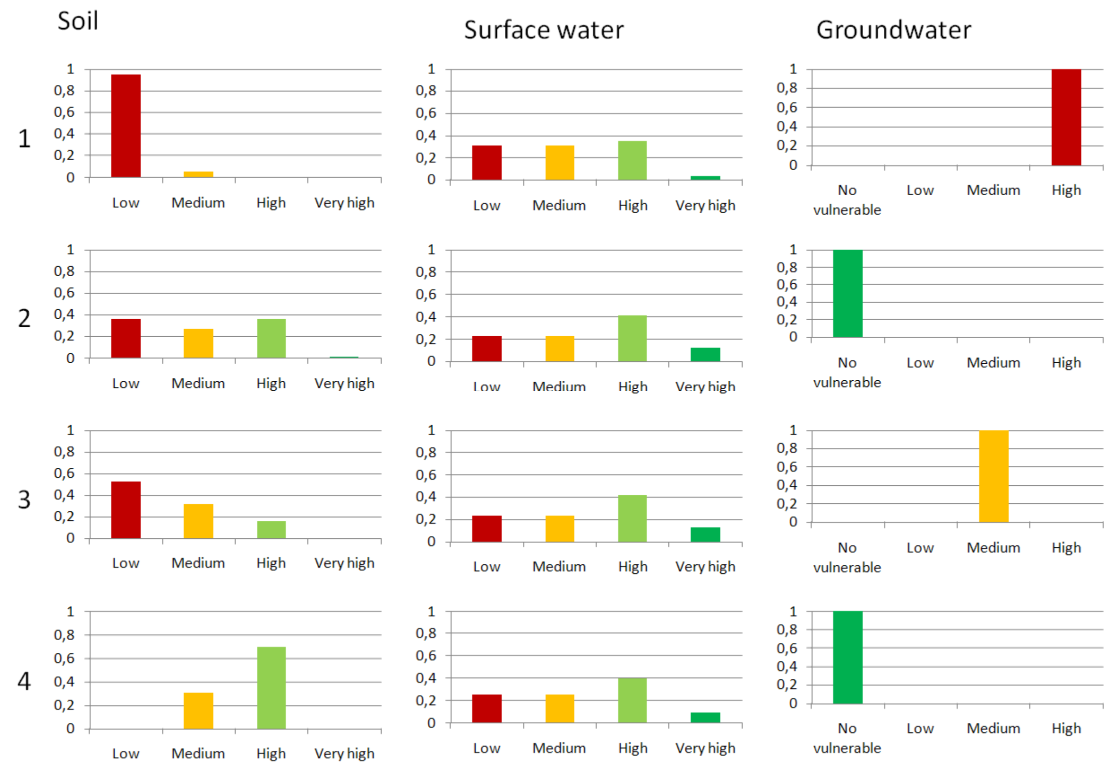

Figure 8.8. CPTs, represented as distributions, for the nodes Soil, Surface water and Groundwater, for the 4 selected cases.

Cases 1 and 3 presented a Low MP value. However, case 3 has a higher uncertainty associated. While case 1 is probably Low (P_{Low}= 0.89), case 3 is likely to be Low (P_{Low}= 0.57), but could also present Medium (P_{Medium}= 0.32) or High (P_{High}= 0.11) classes. Case 1 lower uncertainty is mainly related to the classification obtained for the Environmental node, as both cases have similar results for the Social node.

Cases 2 and 4 also have the same MP class (High). Case 4 also presented a high uncertainty for Suitability, being probably High (P_{High}= 0.55), but possibly Medium (P_{Medium}= 0.31) and unlikely Low (P_{Low}= 0.13). These results reflect the likelihood of being High for the Social (P_{High}= 0.47) and Environmental (P_{High}= 0.60) nodes.

In contrast, area 2 presented an even higher uncertainty for the node Suitability, being High (P_{High}= 0.51) or Very high ($P_{Very\ high}$= 0.48). In this case, the Low classification did not occur, indicating a good suitability level. These results are related to a good

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classification for the Social node (Very high) combined with a high uncertainty for the Environmental node. Figure 8.8 shows the results for the parents of the Environmental node. For case 2, both Soil and Surface water nodes present highly uncertain values, fact that justifies the uncertainty on the child. The Very high classification obtained for the Social node on case 2 was expected as this area present the higher distance to cities.

There is an observed tendency for lower probabilities in the Environmental node (Figure 8.7) if any of the parent nodes (Figure 8.8) has a Low classification. This is a consequence of the method employed to construct the CPT for the Environmental node, which tends to consider the worst possible classification. Also, uncertainty increases when disparate values are observer for the parents (case 2).

4 Conclusions

Sewage sludge management on agricultural soils is an issue of great concern due to population increase and limited availability of area. Land classification allows more confident decisions due to the differentiation of the areas inside the region. The classification is a tool for better management practices, to ensure future sustainability.

The use of BN techniques for land classification presents several advantages when compared to other integration techniques. The most important ones are the ability of dealing with the uncertainty related to model development and the capacity of integrating incomparable and sometimes incommensurate data. The complexity of the system can be represented without the need to integrate processes at different scales, but only at the separate scale of behaviour recognition for each process (Ticehurst et al., 2007).

The probability maps gave an improved perception of systems uncertainty, clearly indicating the areas with more likelihood of reaching any of the defined classes. The geographical representation combined with the evaluation of the CPTs improves the understanding of model uncertainty and may be useful for further evaluations on

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specific agricultural areas. The assessment of the CPTs allows identifying the sources of uncertainty in the model, being coherent with model development.

BN evaluation with experts is also important. This can be done via a structured review of the model. For the suitability model, a review of the results was conducted with experts. Overall, the model received positive feedback. The model was regarded as being a reasonable representation of a complex environment, but the need for routine updating was emphasized.

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Discussion Part II

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The developed Spatial Multicriteria Decision models allowed identifying the best areas to amend with sewage sludge within the studied region of Catalonia (Spain). As previously described, the region has an agricultural area of more than 1 million hectares and an annual SS production of 140,000 tonnes dry weight. Chapter 6 presented a spatial evaluation framework based on the LSP method.

According to the evaluation described in Chapter 7, the suitable areas represent more than 680,000 hectares, being sufficient for managing the amount of sludge produced in Catalonia. The most important agricultural area of Catalonia is located at the central depression. This area was proven to be the best one to amend with sewage sludge, due to its soil and landscape characteristics. The Ebro River catchment area was observed to be also important. In contrast, some areas are considered not suitable for amending with sewage sludge. For these areas, more studies should be performed to define the best types of fertilizer.

Chapter 7 also presented a detailed evaluation of this model. Global sensitivity analysis confirmed that groundwater vulnerability is one important criterion of the analysis. In fact, the points with high groundwater vulnerability were mainly located in areas with low suitability. This criterion explained 23% of the results variability. The distance from the field to urban areas presented also a pronounced importance, explaining 19% of results variability. Metals, crop type and slope were also notably important. These results agreed with the expectations of the stakeholders. In fact, the model has been extensively calibrated in order to achieve these results.

The exploratory evaluation measured the error associated to the exclusion of each map. It was found that the exclusion of crop type map would lead to the higher error on model results, with almost 25% of the pixels having wrong outputs. This result was expected as in the evaluation of the suitability of agricultural fields the crop type should be one of the most important factors of the evaluation. Most of the suitable areas correspond to fruit and cereal fields, while cabbage and cattle fields correspond to areas with low and very low suitability values. The other evaluated scenarios also presented important proportion of error in the outputs (between 6.1 and 9.7%).

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The results of this work are strongly valid to give insights related to the developed spatial model, as they represent an evaluation of the input data. In fact, the results were presented to the stakeholders and most of them agreed with the given outputs. However, the use of multicriteria tools, especially the LSP method, presented an important drawback. Unfortunately, this tool does not incorporate the uncertainty related to model development. Due to this fact, a Bayesian network (BN) was constructed to represent and communicate the uncertainties related to model development.

Chapter 8 provided the results of the application of BNs to the same case-study. The results obtained were highly correlated to those presented on Chapters 6 and 7. The representation of the uncertainty in the model results allowed a better understanding of the results, as it gived the probability of reaching each suitability class. In consequence, more confident decisions could be made as the likelihood of reaching each value is explicitly defined.

One clear example is the central depression area. While the first study presented clearly defined classed with some strong changes between classes in a low distance, BN results provides a broader evaluation of each pixel, clearly pointing out the uncertainty related to each class. The use of CPTs was also proved to help the decision maker to understand the sources of uncertainty for each evaluated pixel.

Future improvements of the models presented in Chapters 6 to 8 might consider changing the evaluation scale, i.e., working in a larger and more detailed scale. In this case, more accurate data could be provided. Moreover, the development of an open tool to present the results to stakeholders could provide more insights related to their understanding of the system.

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Chapter 9

Conclusions

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In this thesis, the problem of managing sewage sludge on agricultural soils was assessed. The evaluation was performed based on environmental modelling and decision support tools.

The first part of the thesis gives a broad evaluation of the fate, transport, exposure and human health risks of contamination due to the presence of persistent organic pollutants (POPs) in the sludge matrix.

- The fate of POPs in the soil matrix was calculated based on fugacity models. The results have proven that the levels reached through a 30 years evaluation are much lower than those reported in the literature for natural soils in the same region, indicating that the atmospheric deposition of these contaminants is a source much more important than the organic amendment. The same tendency was observed in the plant models, i.e., particle deposition and diffusion air-plant are the most important routes, and root transfer plays a minor role in plant uptake of pollutants.

- Regarding the transfer to the cattle, the ingestion of soil was proved to be the most important exposure pathway. In order to reduce cattle exposure, other application types, such as sludge injection, should be considered.

- Concerning human health exposure, the ingestion of milk and meat were found to be the most important routes. Again, the change on the application type could reduce the concentration in the food chain. It must be highlighted that the application of sludge on cattle fields is banned on several countries. In these cases, the exposure values would be much lower than those reported in this thesis.

- The reported values for cancer and non-cancer risks were below the threshold defined by the local environmental agency, leading to the conclusion that the use of biosolids as an organic amendment does not mean any health risks for the population, if the conditions described in this thesis are respected. That is, if the maximum levels of contaminants in sludge, application dose and type, as

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well as the limits defined by the national and European legislation, are regarded.

- Half life in soil, consumption rates of meat and milk, and the amount of soil ingested by cattle are the most sensitive parameters of the environmental models, as shown by the sensitivity analysis results. Model enhancement shall consider a better estimation of these parameters.

The decision support tools presented in the second part of the thesis determined the best areas to amend with sewage sludge. In fact, the BNs and SMCA decision support tools presented the trends for the areas that are more suitable to receive sewage sludge, considering a list of 12 environmental and human health criteria.

- The results of model application to Catalonia indicate that the southern area of the central depression is the best one to amend with sewage sludge. This area presented High and Very High suitability classes in both developed tools.

- Crop type and groundwater vulnerability are the most important parameters in the land classification problem. Distance to urban areas, metal concentration in soil and slope presented also a marked importance.

- The use of decision support tools provided a systematic and intuitive approach to guide the decision making process, considering the best available knowledge, and integrating information provided by different sources. The tools present a structured analysis of the decision process, guided by stakeholders concerns, providing related information, suitable tools to address problems and encouraging discussion of the possible solutions.

- The integration of decision models in GIS eased data gathering, handling and results representation. This type of representation is useful for the decision makers to visualise the quantity and location of suitable agricultural fields, and define how to distribute the amount of sludge produced.

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- The application of Bayesian networks allowed a better understanding of the uncertainty related to model development. The output maps of the analysis represent not only the general trends but also the probability of reaching any of the classes. A high uncertainty was found for most of the areas. A model improvement would reduce the uncertainty related to some points. This could be performed through the development of more workshops and/or elaborating questionnaires to the stakeholders, trying to translate in a more precise way their expectations.

- Some studies in a larger scale could be performed for the most problematic areas, with low suitability values and high related uncertainty. These studies would allow administrators to perform decisions based on the regional production of sludge and its specific characteristics.

As any environmental problem, the management of sewage sludge on agricultural soils evolves a broad number of aspects, such as food safety and health risks. These aspects, combined with the maintenance of soil quality in agricultural areas, lead to different expectancies by the evolved parts. In addition, these issues do not have a common scale to be compared and they are very hard to predict. Consequently, the development of environmental tools for the management of this residue is crucial to maintain safe environmental levels, considering the engagement of the different stakeholders.

Finally, the developed tools for the management of sewage sludge on agricultural soils were proven to successfully tackle this problem. These methodologies are able to represent a new paradigm, where all the stakeholders are involved, and expert knowledge is inserted. Given the urgent need of better environmental management practices, not only for agricultural soils but in other strategic fields, these tools represent a key issue to reach an improved sustainability.

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Annex

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List of publications derived from this PhD thesis

Scientific articles and book chapters

Bojarski, A., Passuello, A., Schuhmacher, M., Jiménez, L. Elaboration and improvement of POPs fate model using sensitivity analysis for parameter selection in sludge amended soils. (under development)

Passuello, A., Bojarski, A., Schuhmacher, M., Jiménez, L., Nadal, M., 2009. Evaluating long-term contamination in soils amended with sewage sludge, in: Athanasiadis, I.N., Mitkas, P.A., Rizzoli, A.E., Gomez, J.M. (Eds.), Information technologies in environmental engineering. Springer-Verlag, Berlin, Germany, pp. 465-477.

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Congress contributions

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Mari, M.; Passuello, A.; Nadal, M.; Schuhmacher, M. Integrated model to assess the risks of amending soils. 20th SETAC Europe Annual Meeting 2010. Seville, Spain.

Passuello, A.; Schuhmacher, M.; Kumar, V.; Nadal, M.; Domingo, J.L. An integrated multimedia model and multicriteria analysis approach to managing sewage sludge application on agricultural soils: framework and methodology. iEMSs - International Congress on Environmental Modelling and Software 2008. Barcelona, Spain.

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Passuello, A., Bojarski, A., Schuhmacher, M., Jiménez, L., Nadal, M. Evaluating long-term contamination in soils amended with sewage sludge. The 4th International Symposium on Information Technologies in Environmental Engineering (ITEE) 2009. Thessaloniki, Greece.

Passuello, A.; Cadiach, O.; Schuhmacher, M.; Pérez, Y. Defining the best agricultural areas to amendment with sewage sludge through a spatial analysis. 20th SETAC Europe Annual Meeting 2010. Seville, Spain.

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Valls, A., Shuhmacher, M., Pijuan, J., Passuello, A., Martí, N. Some approaches to the use of MCDA tools for the management of sewage sludge application on agricultural soils. 69th Meeting of the Europ Working Group Multiple Criteria Decision Aiding 2009. Brusels, Belgium.

Poster presentations

Passuello, A.; Nadal, M., Schuhmacher, M., Domingo, J.L. Assessing the environmental impact of biosolids applied on agricultural soils: fate and exposure of POPs. SETAC North America 29th Annual Meeting 2008. Tampa, USA.

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