A spatially explicit model to calculate Cadmium balances in agro-ecosystems in the EU-27: model description and scenario analysis

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Abstract

Accumulation of Cadmium (Cd) in soil can affect human health and ecosystem functioning once critical levels in crops and soils, respectively, are exceeded. At present, however, an approach to quantify regionally explicit metal accumulation rates at the European scale is lacking. Here we present a regionally explicit model based on 40000 unique combinations of soil type, administrative region, slope class and altitude class that is able to calculate changes in soil Cd accumulation rates, Cd concentrations in soils and crops. Inputs via mineral fertiliser, lime, manure, atmospheric deposition and biosolids (compost, sludge) are based on down-scaled regional (NUTS3) or national data, whereas atmospheric Cd deposition is based on 50 km x 50 km model results. Outputs via leaching and crop uptake were calculated at NCU (Nitrogen Calculation Units) level using transfer models that account for differences in soil metal content, pH, organic matter, and clay content, and taking into account regional differences in water fluxes and crop type. Calculated current Cd balances at the EU-27 level indicate that Cd accumulation rates are -0.49 g Cd ha^{-1} yr^{-1} for grassland (range: -3.1 to +1.7 g Cd ha^{-1} yr^{-1} at country level) and +0.59 g Cd ha^{-1} yr^{-1} for arable land (range -1.1 to + 1.35 g Cd ha^{-1} yr^{-1} at country level). Cadmium leaching from soil appears to be the critical factor that controls the balance of Cd in most areas and the choice of model used to calculate the Cd concentration in solution has a large impact on the overall outcome. Predicted absolute changes in the soil Cd content at country level after 100 years for scenarios using Cd levels in mineral P fertiliser ranging from 0 mg Cd kg^{-1} P_2O_5 to 80 mg Cd kg^{-1} P_2O_5 are relatively small and vary from 0% in case of the Cd-0 scenario to +12% in case of the Cd-60 scenario for arable soils compared to current levels in arable soils. For grassland soils, Cd balances are, at EU level, negative and predicted relative changes in soil Cd levels range from -16% (Cd-0) to -1% (Cd-60) compared to current levels. Dynamic scenario calculations indicate that a maximum level of 20.7 mg Cd kg^{-1} P_2O_5 in mineral P fertilisers is required to achieve an EU-wide (arable + grassland) stand-still, i.e a zero net change of the Cd level in soil after 100 years. For pasture soils, receiving lower amounts of P fertiliser an acceptable level of 64.2 mg Cd kg^{-1} P_2O_5 would lead to stand-still but for arable soils a critical value of 0 mg Cd kg^{-1} P_2O_5 would be required in order to avoid a further build-up of Cd in arable cropping systems.

Introduction

The presence of heavy metals in arable soil and pasture soils and accumulation thereof due to continuous inputs via atmosphere and soil amendments is of concern in view of food safety (EFSA, 2012), water quality (e.g. Peng et al., 2016, Mirzabeygi et al., 2017) and soil health, notably the impact on micro-organisms (e.g. Giller et al., 1998). Accumulation of Cd in food crops and subsequent transfer into the food chain is one of major exposure pathways for animals and human beings (Franz et al., 2009; Peng et al., 2016). Recent estimates by the European Food Safety Authority (EFSA, 2012) indicate that the average exposure for infants and the 95th percentile exposure for adults is in excess of the reduced Tolerable Weekly Intake (TWI) of 2.5 µg/kg b.w. per week, which led EFSA to conclude that there was a need to reduce intake of cadmium via dietary exposure. Since Cd in important food groups like vegetables or grain products are positively correlated to the Cd content in the soil (McLaughlin et al., 2011), reducing levels of Cd in soils via reduction of inputs seems a viable option to reduce exposure. However, the total intake of Cd via food cannot be related entirely to the Cd content in soil since part of the food consumed by people is either not produced in the EU (a.o. intake via chocolate) or has no relation with soil at all. This relates a.o. to Cd intake via seafood or products grown in soil-less cultures including vegetables grown in greenhouses. Nevertheless, recent estimates show that up to 54% of the total Cd intake by average consumers in the EU can be related to the Cd content in soils (Rietra et al., 2017). Hence, reduction of Cd inputs to soil is one of the tools to lower Cd accumulation rates or even achieve a negative balance which ultimately would result in lower levels of Cd in soil. At present however, it is unclear how fast and to what degree a reduction of Cd levels in soil can be obtained so as to reduce exposure of Cd below the TWI. Recent inventories in Switzerland (Bigalke et al., 2017) and New Zealand (Schipper et al., 2017) for example showed that inputs of Cd inputs to arable cropping systems have been reduced, but the response of the soil Cd level to such reduced inputs is slow or absent.

Accumulation of Cd in soil occurs when inputs to soil exceed outputs. Major sources of Cd include the use of mineral P-fertiliser and animal manure, inputs via atmospheric deposition and the use of organic soil amendments like compost and sludge (Amlinger, 2004; Nicholson et al., 1999, Eckel et al., 2005). During the last two decades, however, industrial emissions, including traffic and waste combustion have decreased substantially (EEA, 2017). Also consumption rates of mineral P fertilizer have decreased from almost 8000 kton in 1980 to 2000 kton in 2010 (Six and Smolders, 2014) which has resulted in a marked decrease in inputs of Cd via fertiliser to soil. Based on the average Cd content in mineral P fertilisers of approx. 40 mg Cd/kg P2O5 (Nziguheba and Smolders, 2008) the reduced consumption rate corresponds with a decrease of the Cd load to arable soil of 320 tons at EU level. Currently, however, there is a trend towards a more efficient re-use of existing resources for nutrients (End of Waste; COM 2005) which includes the use of organic resources like compost, digestate, biochar or sludge in agriculture to replace part of the mineral fertilisers used at present. Such resources can contain elevated levels of contaminants and the partial replacement of mineral fertiliser by such organic resources may induce a higher load of contaminants to agro-ecosystems across the EU. In addition, proposals to reduce the minimum nutrient requirements for fertilisers (COM, 2016) also can lead to an increase in the load of contaminants present in such products. The degree to which such policy changes affect the regional or national load of contaminants including Cd to soil is, as of now, unclear although estimates at country level for the Netherlands suggest that loads of Cd can increase from the current load of 2.6 tons Cd/year to 7.2 or even 15.9 tons/year if all mineral fertiliser applied were to meet the minimum nutritional requirements (Römkens et al., 2016).

Accumulation of Cd has been extensively documented both at farm level (Eckel, 2005), regional and national level like including but not limited to studies for the Netherlands (de Vries et al., 2004); Switzerland (Keller and Schulin, 2003) and the UK (Nicholson et al., 2006). On a local scale numerous case studies have been performed showing that in arable cropping systems inputs exceed outputs (e.g. Moolenaar and Lexmond, 1998; Keller and Schulin, 2003; resulting in a substantial accumulation of Cd in soil with time. Recent studies however also document a potential decline in accumulation rates either due to reduced inputs or increased removal via leaching (Salmanzadeh et al., 2017). However, the majority of studies including an EU-wide compilation of farm and field metal balances by Eckel et al (2005) show that metal accumulation is common but accumulation rates vary substantially between countries and farm type. Typical (median) accumulation rates for Cd are +1.7 g ha^{-1} yr^{-1} but the ranges are substantial and vary from -0.3 g ha^{-1} yr^{-1} to +20 g ha^{-1} yr^{-1}. More recent estimates of Cd balances at EU level using representative average data from soils and Cd inputs and outputs also suggest that Cd balances at the EU level are close to equilibrium and predicted changes in the Cd content of soils range from -64% to +12% relative to the current level within 100 years from now (Six and Smolders, 2014). The latter results however are not based on real combinations of soil, land use and location but based on simulated distributions of soil properties and variable inputs. The distribution rates used, however are rather representative for the range in Cd levels in soils and soil properties.

Despite the range in observations of the degree of accumulation of Cd in specific arable cropping systems and the recent concern about current exposure levels due to intake of food (EFSA, 2012) most European agricultural soils still can be considered as relatively uncontaminated (Reimann et al., 2014) and the quality of food, ecosystem and water quality at large is not yet under threat (de Vries et al., 2007). This obviously does not apply to areas with high actual or historic inputs from industry. Such areas include for example areas near (former) mining sites (e.g. Van der Fels-Klerkx, 2011; Rodrigues et al., 2012) where levels in animal feed exceed current feed quality standards. This shows that there is a clear need for a regionally explicit tool to assess where Cd levels in soil can lead to excess exposure or Cd balances will remain positive thus leading to a further increase of the levels in soils.

At present, a model approach to calculate and compare metal balances and the impact of land use or proposed policy changes like the proposed revision of the EU Fertiliser Regulation (EU2003-2003) on soil and crop quality at a *regional* level across the EU, is still lacking. This is partly due to the fact that it proved to be quite difficult to obtain reliable data on the major components of the metal balance including both inputs and outputs (Eckel et al., 2005). Key issues to be addressed are a.o. the derivation a consistent set of input data related to levels of metals in various sources like manure, sludge and inorganic fertilizer across many different types of land use, climate soil type etc. Due to the variability in both application rates and quality of soil amendments like manure and compost direct estimates of inputs to soil and outputs from soil regarding metals are lacking in many countries across the EU. One of the first approaches to obtain estimates of accumulation rates and long term changes in soil quality on a national level has been developed for Canada (Sheppard et al., 2009). As such this is a first approach applied at this scale level but also here, the approach was based on a rather limited number of soil data and inputs of manure were related to contaminant levels in feed which is not necessarily representative for manure considering the wide range in metals in different feed compounds (Nicholson et al., 1999). To overcome this limitation, fluxes of metals to and from soils have to be derived partially from meta-information such as nutrient loading rates that can be converted to equivalent loading rates of metals. In this study we will use such data which are available at the regional level to convert fluxes of N and P to soil to fluxes of metals. This can be done based on available data regarding the metal to N or P ratio in the major inputs to soil including manure, fertilizer and sludge.

Aside from more precise estimates of inputs, also outputs from soil including leaching and crop uptake are often poorly quantified or based on average data. Even though leaching may not be equally relevant in all agro-ecosystems, in areas with high groundwater tables leaching of metals from soil to ground- and surface waters is one of the major pathways by which metals are released into the environment. Model studies by Moolenaar and Lexmond (1998) and Bonten et al. (2012) for the Netherlands indicate that leaching makes up for 70% of the total Cd outputs from soil (aside from crop off-take). Estimates at EU level (Six and Smolders, 2014) even suggest that leaching losses are equal to more than 90% of the total Cd output from soil. Despite the obvious relevance of leaching to construct a reliable Cd mass balance, Cd balances do not always account for leaching partially because reliable estimates of leaching fluxes are difficult to obtain. Hence, in most studies presenting Cd balances, models are used to estimate leaching losses. Clearly reliable estimates of leaching losses based on model predictions are prone to model uncertainty but during recent years various robust Cd partition models have been developed and calibrated that allow the user to calculate soil solution concentration based on a combination of soil properties including pH, organic matter and clay content (a.o. Groenenberg et al., 2012; Six and Smolders, 2014). Such models appeared to give reliable results over a wide range of soils when applied to independent data. The same is true for crop uptake models and even though the contribution of crop uptake appears to be quantitatively smaller compared to leaching losses, relatively simple transfer models are now available and despite the variability between regions and crop varieties, such models allow for a reasonable estimate of crop off take rates of Cd from soil (McLaughlin et al., 2011)

The aim of this paper is to describe the framework of a spatially explicit model at EU-27 level that is able to calculate Cd metal balances at a regional level considering all relevant in- and outputs based on local, regional or national data in combination with models to calculate leaching and crop uptake rates. The model will be used to calculate current Cd balances at a regional level which will be up-scaled to the national and EU level. Aside from the current balance ("Business as Usual", BaU) current Cd balances will be presented for grassland and arable land as well as Cd balances that reveal the impact of a range in Cd levels in mineral P fertilisers in line with the proposed Cd limits for mineral fertilisers (EU2003-2003). The impact

2 Material and Methods

2.1 Model Outline

To calculate Cd balances for the topsoil in agro-ecosystems the INTEGRATOR model is (De Vries et al., 2011a, b; Kros et al., 2012) is used. INTEGRATOR is an environmental agricultural model which calculates nitrogen and greenhouse gas emissions from housing and manure storage systems, agricultural soils, non-agricultural soils and surface waters for the EU-27 (note that for metals no data for Malta and Cyprus were included, results are presented for 25 Member States only). The model was developed to calculate detailed nutrient (N, P, K) balances at NCU and NUTS3 level considering a.o. inputs via (animal) manure, mineral fertiliser, lime, biosolids and atmospheric deposition as well as outputs including leaching losses, and crop removal rates. In addition some specific processes like volatilization and nitrogen fixation are accounted for in INTEGRATOR (de Vries et al., 2011*a,b*; Kros et al., 2012) but these are not considered relevant to calculate Cd mass balances. In addition, data for

Malta and Cyprus have been omitted since these were largely not available. Equation 1 shows the fluxes that are considered in the Cd balance, including both inputs and outputs:

$$F\text{-}Me_{.} = F\text{-}Me_{man.} + F\text{-}Me_{inorgfert..} + F\text{-}Me_{lime} + F\text{-}Me_{biosolids\ .} + F\text{-}Me_{atm\ dep.} - F\text{-}Me_{plup} - F\text{-}Me_{leach} \quad [1]$$

With

F-Me:	Cadmium flux in g ha^{-1} yr^{-1}
Man.	Sum of cattle, pig and poultry manure
Inorgfert	Sum of mineral N, P and K fertilizers
Lime	Sum of lime and dolomite gift
biosolids	Total of compost and sewage treatment plant derived sludge
Atm.Dep	Atmospheric deposition
Pl.up.	Plant uptake
Leach	Leaching loss from topsoils (0-10cm for grassland, 0-30cm for arable land)

A schematic representation is given in figure 1, this includes also an overview of the scale at which various data are available which either requires down- or upscaling.

Here, we only consider inputs and outputs to and from the topsoil, both for grassland and arable land. In case of grassland a depth of 10 cm was used, in arable soils a layer of 30 cm was used. These layers are assumed to be the most relevant in view of crop quality in that the majority of the uptake from soil will occur from the 0-10 cm layer (grassland) and the 0 – 30 cm (arable land). To calculate metal fluxes for all inputs from fertiliser, manure, and biosolids, the calculated nutrient fluxes for N, P and K (INTEGRATOR) are converted to corresponding metal fluxes. This includes inputs from mineral fertiliser (for N, P, and K), animal manure (cow, pig and poultry), bio-solids (compost and sludge)

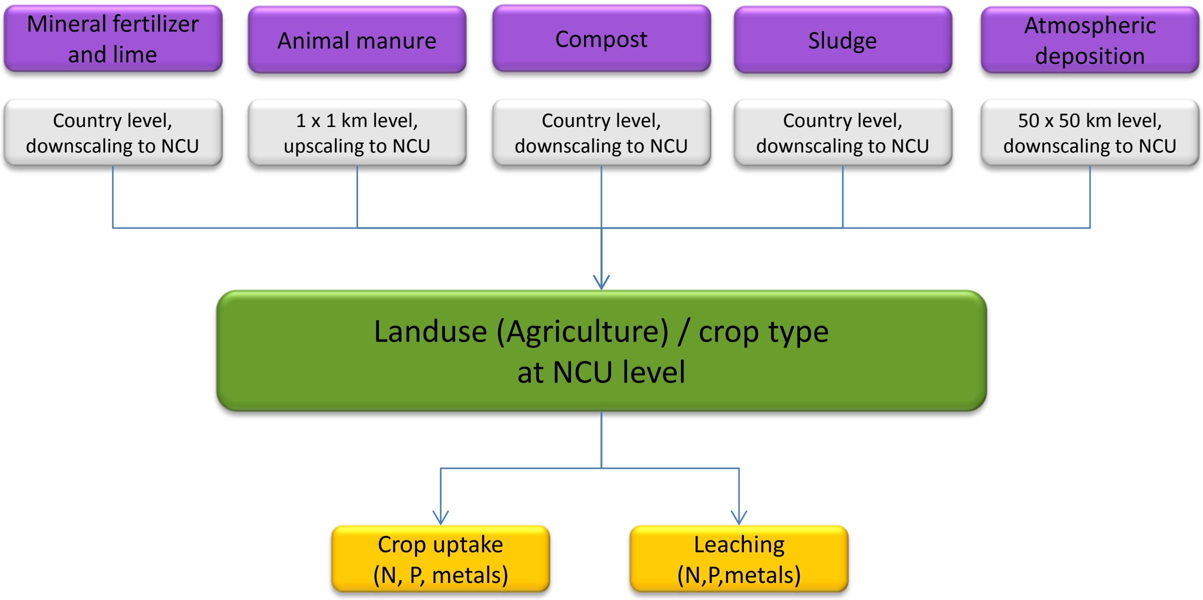


Figure 1. Main components considered in the mass balance model and scale level of original data

Inputs of metals by manure, sludge, fertilizer were calculated as the product of the load of the product in kg N, P or K per hectare per year and the metal to N, P or K ratio of the specific product. This approach was chosen since application rates for N, P and K are available at the desired scale level by the INTEGRATOR model and we thus only needed to convert N and P fluxes to metal fluxes. The Cd-load from application of lime was calculated based on the total consumption rate of lime (data at country level) times the average cd content in lime. Data on atmospheric deposition were obtained from Ilyin et al. (2009) which provides regionally explicit inputs at a 50x50 km grid. Metal removal rates via crop off-take where was calculated via the crop yields based on the crops specified in the CAPRI (Britz and Witzke, 2008) database (removal rates in ton dry matter ha^{-1}) which were converted to equivalent removal Cd rates using appropriate Uptake Factors for specific crops or crop groups. Leaching losses are calculated using the net water flux at the bottom of the layer considered (i.e. -10 or -30 cm) calculated as the difference between rainfall and crop specific evapotranspiration rates which are then multiplied by the calculated concentration of Cd in the corresponding layer considering differences in the Cd content of the soil, pH, organic matter content and clay content.

2.2 Basic data used at NCU level and calculation of nutrient fluxes

2.2.1 Spatially explicit data used in Integrator

Input data include representative soil properties at NCU level. For each spatial unit (NCU), representative values of organic matter, clay and pH-$CaCl_2$ are based on the data in the Soilgrids databases (Heuvelink et al., 2016). A recently developed machine learning approach (Hengl et al., 2014 and Heuvelink et al., 2016) was used to derive 1x1 km data using the source data listed after which the median value of all grid cells within a specific NCU was used as representative value for the NCU as a whole. The Corine landuse map (EEA, 2009) was used to distinguish between arable land and grassland to derive separate values for pH, organic matter and clay for both types of land use. For Cd especially the significant difference in pH between arable land and grassland is relevant in view of the impact of pH on the Cd solution concentration which serves as a basis for the calculation of leaching losses.

Total Cd levels in soils at a 1x1 km level applying the same technique have been obtained using the data from the GEMAS (Geochemical Mapping of Agricultural and Grazing Land in Europe, Reiman et al., 2014) database. Considering the relatively low sample density in this database, the data from grassland and arable land were used together to derive the best fit model. At NCU level a single value for Cd was derived using the best fit model at a 1 x 1 km grid level. The median value of all 1x1 km grids within a specific NCU was subsequently used as representative level for Cd at NCU level. Atmospheric metal deposition (in g ha^{-1} yr^{-1}) is derived by downscaling results from the EMEP heavy metal (HM) model (Ilyin et al., 2009) available in a 50 km x 50 km grid to NCU level.

2.2.2 Calculation of manure, mineral fertilizer, biosolid and lime gift

The total N and P loads via manure, mineral fertiliser, biosolids and lime were calculated based on *i*. crop nutrient demands, *ii.* total allowed manure application rates according to EU legislation (COM, 1991), *iii.* animal density and manure production per animal, *iv.* total country wide consumption rates of mineral fertiliser and biosolids and, *v*. soil pH for lime application (de Vries et al., 2011 a,b). Here we describe how inputs for the various fluxes were derived using country specific of EU-wide data depending on the availability and assumed variability between countries.

Calculation of application rates of manure, fertiliser, and biosolids

Initially the N and P load via manure is calculated based on the data on animal density and allowed manure application. If the amount of available manure exceeds the required amount, the excess manure is distributed in surrounding spatial units. Data on animal density, crop demand, crop type and soil type are available at the NCU level (Neumann et al., 2009; Kros et al., 2012) and hence used to calculate the manure gift at NCU level. The remaining mineral fertiliser application rate, for which no data are available at NCU level, then are calculated as the difference between recommended gifts which are crop and soil specific and the gift supplied via manure. This is done separately for N and P. Subsequently the total calculated mineral fertiliser gift, defined as the sum of all gifts at NCU per country, are scaled to the known national data to ensure that the total fertiliser consumption is in line with known data (de Vries et al., 2018). For both compost and sludge no spatially explicit application data are available and country wide consumption data, separate for compost and sludge, by agriculture are used (Barth et al., 2008; Evans, 2012) which are equally applied to the arable land fraction within the NCUs that is used for fodder production (maize) and arable crops with a medium to high N demand; no biosolids are applied to grassland. The equal distribution of biosolids across all arable land clearly is an oversimplification but the error thus introduced on the metal balance is small due to the small amount of biosolids used compared to manure and mineral fertiliser. For lime only country level consumption data are available (UNFCCC, 2012) and this amount is equally divided, at country level, among all arable land with a pH < 6.5. No distinction between Dolomite and lime was made and a single composition was used (Dittrich and klose, 2008) to calculate the resulting Cd inputs. With the exception of the UK and Ireland, lime was applied to arable land only.

In table 1 an overview of the source data used in this study is given including the data that were used to calculate the corresponding metal load that results from the application of the fertilisers, biosolids and lime applied here.

Table 1 overview of source data and conversion used to derive country or EU specific levels of Cd in inputs

Compound	subdivision	source	Conversion applied
Animal manure	Cattle slurry, cattle manure, pig slurry, pig manure, poultry manure	Amlinger, 2004. Annex 4 table A4.3	Country specific data were compiled and based on number of samples per country, an EU mean value was derived. Levels in poultry manure were used for other manure categories (notably manure from fur animals)
Mineral fertiliser	N fertiliser incl. ammoniumsulfaat, ureum, Kalkammonsalpeter, ammoniumsulfaat ureum, zwavelzure ammonia	Dittrich and Klose 2008	Values as reported have been used for all countries
	P fertiliser	Smolders, 2017	Country based average value have been applied based on data from countries included in the study
	K fertiliser incl. KCl 60,	Boysen (1992)	Values as reported have

	K_2SO_4, patentkali30%	Dittrich & Klose (2008)	been used for all countries
Liming Materials	No subdivision between products aplpied	Dittrich and klose (2008)	Total lime gift at country level is equally distributed to arable land with pH < 6.5
Compost	Data on production andd applicaiton of Green Waste, Biowaste and mixed compost are used; sludge derived compost was excluded	ORBIT 2008 Amlinger (2004) Barth et al. (2008) Lesschen et al. (2013)	Country specific cd content in compost was used for those countries that reported data, EU average was used for all other countries
Sludge	No specific products were distinguished	Evans (2012) JRC (2012) EC (2010)	Country specific cd content in sludge was used for those countries that reported data, EU average was used for all other countries

Source data used to calculate the metal load from fertilisers, lime, compost, sludge and animal manure

Country average values for the Cd content in P fertilisers have used derived from the study by Smolders (2017). In total 389 samples of P fertilisers with a P_2O_5 content of 5% or more are included in the database. Country wide average levels of Cd expressed as mg Cd kg^{-1} P_2O_5 range from 0.7 to 58.1 mg Cd kg^{-1} P_2O_5. For countries without data a value of 32 mg Cd kg^{-1} P_2O_5 was used which is equivalent to the weighted mean value correcting for the actual use of P fertilisers in the countries included in the study (Smolders, 2017). For nitrogen and potassium fertilisers EU wide average data have been used as reported by Dittrich and Klose (2008) and Boysen (1992) without further specification. For lime, data from Dittrich and Klose (2008) were used for all countries to account for inputs of Cd via lime. To calculate country specific application rates for compost, data from ORBIT (2008) were used on country specific production rates and application rates in agriculture. Here we used data on Green compost (GC), Biowaste compost (BC) and Mixed compost (MC). Sludge based compost was not included since inputs from sludge are accounted for directly and double counting was thus avoided. Data from Amlinger (2004) and Barth et al. (2008) were used to calculate an EU-wide median cadmium level in each of these three types of compost after correction for the number of samples collected in each country. Recent data published by JRC (2014) indicate that average levels of metals in various types of compost largely remained the same as those published by Amlinger (2004). Data of the metal content for mixed waste compost are not available and the average of green waste and biowaste compost was used instead. For all countries the median value of all reported data has been used. Multiplication of the total compost application rate at country level of compost by the averaged Cd content in the three types of compost yields the total Cd load at country level. To calculate the country average Cd to P or N ratio, data from Lesschen (2013) were used who reported the average N and P content in GC, BC and MC. For those countries not included in the study by Amlinger (2004), the median value of all countries was used. Sludge production and application data at country level were taken from Evans (2012). The Cd content as well as the N and P content in sludge was based on country specific data as listed in the Working Document on Sludge and Biowaste (EC, 2010). For countries with no reported data the median value of data as reported by JRC (2012) was used. Data for manure at country level can vary substantially, even within countries. Here data for slurry (cattle and pig slurry) and solid manure (from cattle, pigs and poultry) were taken from Amlinger (2004) being a representative set of data at EU level. For each of the 5 types of manure distinguished, EU average values were derived and applied to all calculation units.

2.2.3 Hydrology

Water fluxes to and from soils were based on the approach from Keuskamp et al. (2012); this allows for the calculation of total runoff, surface runoff, and the excess water flowing recharging shallow groundwater at a 1 x 1 km grid level. Differences in climate, rainfall, soil type, lithology, land use and irrigation are taken into account.

2.3 Model inputs and conversions used to calculate cadmium fluxes from inputs considered

2.3.1 Animal manure.

Cadmium inputs via manure (in g/NCU) are calculated by multiplication of N excretion rates at NCU level (in kg/NCU) by the Me/N ratio of manure for the 5 types of manure for which data are available:

$$[Me/N]_{manure} = Me\ content_{manure}/N\ content_{manure} \qquad [2]$$

$$Me_{load\text{-}NCU} = N\text{-}excretion \times Me/N\ ratio_{manure} \qquad [3]$$

The Me/N (g_{Me}/kg N) ratio is derived for all eight types of manure used in Integrator by division of the metal content in manure (in mg_{Me}/kg dry matter) by the N content in manure (in g N/kg manure dry matter). In total eight types of manure (similar to those used to calculate N and P application rates for manure) are considered, including solid or liquid (slurry) manure. However, data for metal concentrations in manure from horses, sheep/goats and fur animals are largely missing. To account for this, data for cattle were used for sheep/goat/horses and data for poultry were used for fur animals. Even though it is likely that levels of metals in manure may differ between regions and/or countries, a single set of values for each type of manure has been used for all NCU's included in the calculations.

2.3.2 Mineral fertiliser.

Metal inputs by fertilizer (kg/NCU) are calculated by multiplying N, P and K fertilizer inputs (at NCU level (kton N/NCU) with corresponding Me/N, Me/P and Me/K ratios (kg_{Me}/kton N, equivalent to mg_{Me}/kg N) in the various fertilizers:

$$Me_{fertilizer} = N_{fertilizer} \times Me/N + P_{fertilizer} \times Me/P + K_{fertilizer} \times Me/K \qquad [4]$$

The Nutrient to metal ratios have been used for 17 main types of fertilizers as used within Integrator (data in Table S1) including single nutrient as well as complex (N,P,K) fertilizers. In case of complex fertilizers, double counting was avoided by using the dominant nutrient for such complex fertilisers only to calculate the corresponding metal load.

2.3.3 Biosolids

Cadmium inputs by biosolids (here we consider only sludge and compost in kg/NCU) are calculated by multiplying application rates of biosolids at NCU level (kton/NCU) with metal contents in biosolids (mg Me/kg biosolid equivalent to kg Me/kton biosolid):

$$Me_{biosolid\text{-}NCU} = N\text{-}load_{NCU} \times [Me/N]_{biosolids} \qquad [5]$$

This was done separately for compost and sludge.

For both compost and sludge the total reported N or P load via either compost or sludge in combination with the total metal load was used to derive a country average Cd to N or P ratio. This ratio was subsequently used in the scenario calculations at NCU level.

2.3 Model Outputs and conversions used to calculate cadmium fluxes

2.3.1 Leaching

Leaching of Cd from the upper soil layer to the deeper horizons is calculated according to:

$$Cd_{Le} = Q_{le,dc} \cdot [Cd]_{ss}/1000 \quad [6]$$

where Cd_{le} is Cd leaching rate from the topsoil (g ha^{-1} yr^{-1}), $Q_{le,dc}$ is water flux leaving the topsoil (depth of cultivation, dc in m^3 ha^{-1} yr^{-1}) and $[Cd]_{ss}$ is the annual average total Cd concentration in soil solution (mg m^{-3} or µg l^{-1}). To calculate the dissolved Cd concentration in solution a non-linear model was used similar to that developed by a.o. Tiktak et al. (1998), Elzinga et al. (1999), and Keller et al. (2001). Solution concentrations in a wide range of soils were obtained by a 1:10 (w:v) extraction using 0.002 and 0.01 M $CaCl_2$ solutions (Römkens et al., 2004) which appear to mimic in situ soil solution concentrations of Cd (de Greyse et al., 2003). In these models Cd_{ss}, is predicted from the reactive soil metal content, Cd_{re}, that represents the total reversibly adsorbed Cd pool in soils (Groenenberg et al., 2017), when accounting for differences in organic matter (OM, in %), clay (%), pH_{CaCl2} (measured in 0.01 M) and DOC (Dissolved Organic Carbon in mg C/L) according to (Römkens et al., 2004):

$$^{10}\log Cd_{ss} = 4.91 + 1.27 \cdot {}^{10}\log[Cd_{re}] - 0.73 \cdot {}^{10}\log[OM] - 0.48 \cdot {}^{10}\log[clay] - 0.39 \cdot pH_{CaCl2} + 0.08 \cdot {}^{10}\log[DOC] \quad [7]$$

With $Cd_{ss\ soil}$ solution Cd (mmol l^{-1}), Cd_{re} the reactive Cd pool (mol kg^{-1}),

Estimates of DOC can be obtained using an empirical regression model based on organic matter and pH, according to (Römkens et al., 2004):

$$\mathrm{Log}[DOC] = 2.04 + 0.73 \cdot \log[SOM] - 0.17 \cdot pH_{CaCl2} \quad [8]$$

The reactive Cd pool is related to the total soil metal content, $Cd_{tot,soil}$, correcting for the soil organic matter content (SOM) and clay content according to (Römkens et al., 2004):

$$\log Cd_{re} = -0.089 + 1.075 \cdot \log[Cd_{tot,soil}] + 0.022 \cdot \log[SOM] - 0.062 \cdot \log[clay] \quad [9]$$

2.3.2 Crop uptake

Removal of Cd in crops is calculated as the product of harvested biomass times the predicted Cd content in crops according to:

$$Cd_{off} = Y * DM_{crop} \cdot Cd_{crop} \quad [10]$$

where Y is the harvested yield (ton ha^{-1} fresh weight), DM_{crop} is dry matter content of the crops (-) and Cd_{crop} is the calculated concentration of Cd in the harvested crop (mg kg^{-1} dry weight). Crop yields as well as the list of crops considered are taken from the CAPRI model (Britz and Witzke, 2008). The concentration of Cd in crops (Cd_{crop} in mg/kg dry matter) is calculated using a linear soil to plant transfer coefficient according to:

$$Cd_{crop} = BCF_{crop} \cdot Cd_{tot,soil} \quad [11]$$

Where $Cd_{tot,soil}$ is total Cd concentration in soil (mg kg^{-1} dry matter of soil) and BCF_{crop} are crop-specific bioconcentration factors (BCFs), relating the Cd content in crops to those in the soil.

Crop-specific bioconcentration factors (BCFs) for all distinguished crops including potatoes, sugar beets, other root crops; vegetables, barley, soft wheat, durum wheat, rye, oats, grain maize, rice, other cereals including triticale; sunflower, olives, oil crops (including rapeseed), citrus, grapes and other crops, were based on Lübben and Sauerbeck (1991); Versluijs and Otte (2001); Smolders et al., (2007) and Römkens et al. (2008, 2009) as listed in table S2 in the supporting information. In reality, the relationship between Cd concentrations in crop and soil depends on soil properties such as pH, clay content and organic matter contents (see e.g. Brus et al., 2002; De Vries et al., 2007b; Römkens et al., 2009, McLaughlin et al., 2011), but such relationships could not be derived for all crops and consequently, simple linear relationships were assumed in agreement with other studies (Six and Smolders, 2014, Smolders. 2017).

Outputs and Scenarios included in the study

The aim of this study is to provide a spatially explicit overview of the Cd balance in current agro-ecosystems. This includes the balance based on the current inputs and outputs, here called Business as Usual (BaU). Aside from the current balance this paper aims to provide insight in the impact of the proposed revision of EU2003/2003 regarding the quality of mineral fertiliser and other soil amendments, Revision of the Fertiliser Regulation (EU2003/2003) among others includes a proposal for a step-wise reduction of the maximum Cd content in mineral P fertilisers (Pcontent > 5%) from 60 mg Cd kg^{-1} P_2O_5 after approval of the proposal to 40 mg Cd kg^{-1} P_2O_5 3 years after implementation of the proposal and 20 mg Cd kg^{-1} P_2O_5 12 years after implementation. Here we will include these 3 levels as separate scenarios (Cd20, Cd40 and Cd60) as well as two additional scenarios; one being a maximum allowed content of 80 mg Cd kg^{-1} P_2O_5, a value being discussed by several stakeholders, the second one being a scenario without additional inputs of Cd via P fertilisers which serves as a reference to establish the contribution of P-fertilisers relative to the other scenarios.

Cadmium balances for all scenarios will be calculated at NCU level and up-scaled to country and EU level to provide insight in the degree of accumulation, or depletion. As such the degree of accumulation or depletion which is the result of the Cd balance does not provide insight in the changes in the Cd content in soil with time. One of the key issues regarding Cd is however the question to what extent the proposed policy changes will affect Cd levels in soil. Aside from the current balance at time 0 (2017) and those related to proposed policy changes, long term (after 100 years) changes in the Cd content in soil will be calculated at NCU level.

Table 2 Summary of scenarios included in the model study

Scenario	Description
BaU	Business as Usual, current inputs as defined by Integrator
Cd0[1]	Inputs from mineral P fertiliser reduced to zero
Cd20[1]	Level of Cd in mineral P fertiliser set at 20 mg Cd kg^{-1} P_2O_5
Cd40[1]	Level of Cd in mineral P fertiliser set at 40 mg Cd kg^{-1} P_2O_5
Cd60[1]	Level of Cd in mineral P fertiliser set at 60 mg Cd kg^{-1} P_2O_5
Cd80[1]	Level of Cd in mineral P fertiliser set at 80 mg Cd kg^{-1} P_2O_5

[1] all inputs except those from mineral P fertiliser are similar to BaU scenario

3 Current Cadmium Balances in European Cropping Systems

3.1 Soil properties and Cd levels in soil

In figure 2 the resulting maps containing input data for the calculations are shown for cadmium, pH-$CaCl_2$, Organic Carbon content and clay content as calculated at the 1 x 1 km level. Here only those areas are shown that are in use for arable crop production or managed grassland. Natural grassland not used for crop or fodder production or natural areas not receiving manure or fertiliser as well as mountainous areas are not included. In Table 3 a summary of the soil properties is given based on the aggregated distribution at NCU level for arable and grassland separately.

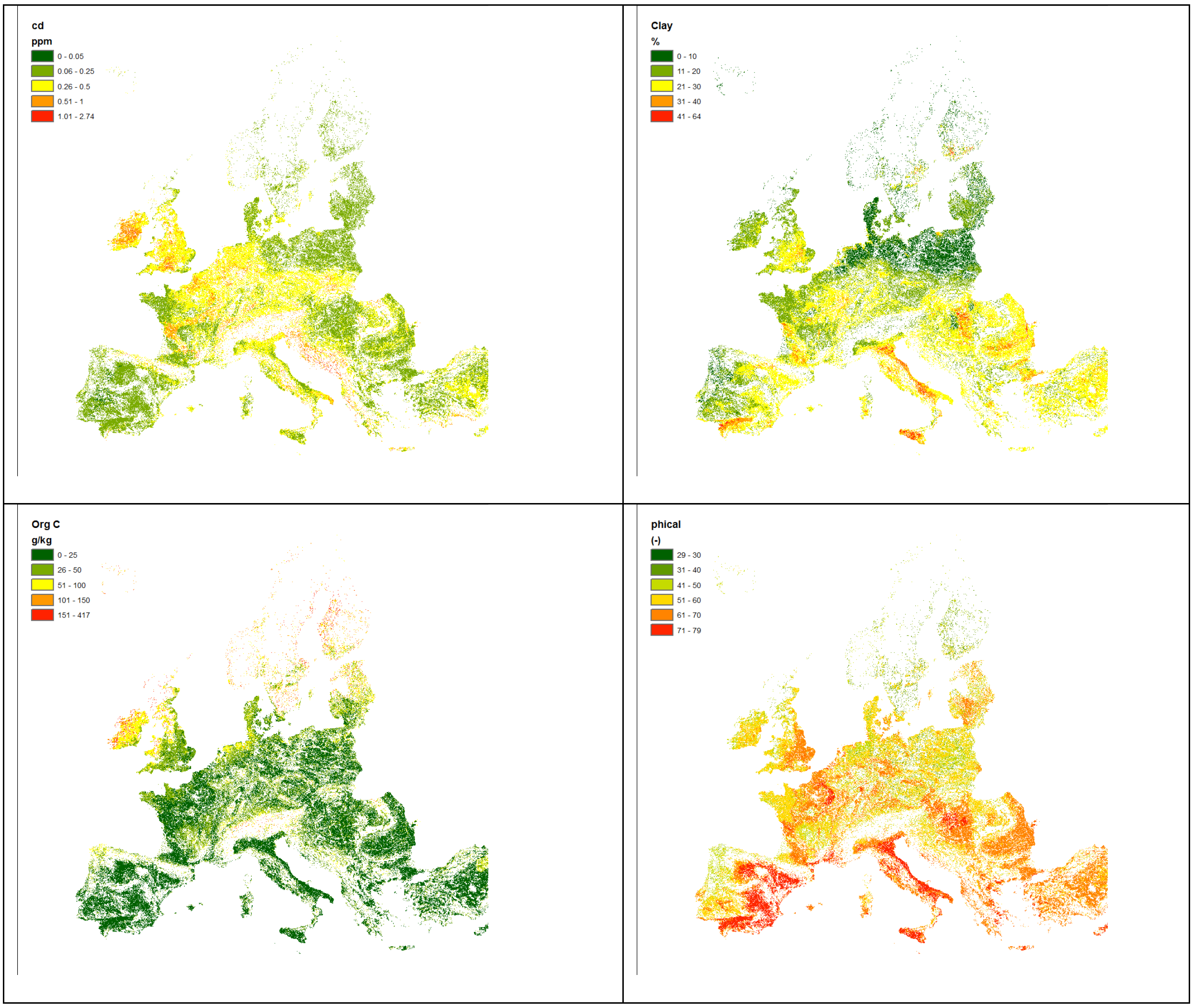


Figure 2 Overview of spatial distribution of Cadmium in the topsoil, clay content, soil organic Carbon content and pH CaCl2 used in the Integrator model.

Table 3. Overview of regionally explicit soil data used at NCU level

	percentile	Area (ha)	Cd soil (mg kg^{-1})	pH $CaCl_2$	SOM %	Clay % < 2 µm	Net water flux mm yr^{-1}
Grassland Soils	min	1	0.04	4.1	0.9	6	25
	5	35	0.14	4.9	2.3	12	45
	25	105	0.23	5.4	3.5	18	174
	50	355	0.30	5.8	4.5	22	259
	75	1289	0.40	6.3	5.9	26	366
	95	7147	0.57	7.0	12.5	35	663
	100	163353	1.29	7.7	100.0	57	1362
Arable Soils	min	36	0.03	4.2	1.0	3	25
	5	79	0.12	5.2	1.4	9	33
	25	218	0.22	5.8	1.9	19	146
	50	864	0.27	6.2	2.4	22	216
	75	3694	0.36	6.7	3.0	28	300

95	23344	0.51	7.3	5.4	37	502
max	318586	1.36	7.7	81.7	60	1141

Data in table 3 show that the range in Cd levels in soils range from < 0.1 mg kg^{-1} to approx. 1.3 mg kg^{-1} in both arable and pasture soils. The median value of 0.3 (arable) – 0.36 (grassland) soils reflects the impact of higher organic matter levels in grassland soils which leads to slightly higher estimates of Cd in pasture soils. Soils rich in organic matter (>10%) are largely found in north-western parts of the EU including the peat soils commonly found in ao Ireland, Scandinavia and parts of Denmark, the Netherlands and Germany. Median pH levels range from 5.8 in pasture soils to 6.2 in arable soils and represent normal values for such forms of landuse. In general pH levels in soils tend to be higher in the calcareous soils in the Mediterranean soils compared to the more acidic soils in the north-western parts of the EU. On average EU soils are characterized by a medium high clay content even though there is a clear distinction between the more sandy soils in the north-western parts of Europe versus the clayey soils that dominate the Mediterranean area. The resulting map of Cd levels in soil reflects a mixture of both history of pollution, elevated background levels, soil type and land use. Relatively high Cd levels are observed in a.o. the Netherlands, Belgium and the UK which is largely related to diffuse and point source pollution. Elevated levels in a.o. Croatia are largely related to elevated background levels in the rocks from which the soils are formed. Low levels of Cd are found in the Scandinavian countries due to a combination of low emission, low soil pH and high rainfall which favours the leaching of Cd from the topsoil. Soil Cd levels are also low in Portugal and Spain which reflect both low background levels and low historic emission from industry or traffic. Data in Table 3 also reveal that there is a large range in surface area covered by the NCU areas, ranging from 1 ha to more than 300000 ha. In order to calculate country or even EU wide representative average values we therefore present such data after correction for surface area.

3.2 Current Cadmium balances in Agroecosystems at country and EU level

Current surface weighted EU average balances for Cd are slightly positive in arable soil (+0.59 g Cd ha^{-1} yr^{-1}) and slightly negative for pasture soils (-0.49 g Cd ha^{-1} yr^{-1}). Due to the larger are used for arable soils compared to pasture, the overall net Ca balance is positive which indicates that at present, Cd is still accumulating in soils. Positive balances for Cd in arable cropping systems and/or associated increases in Cd levels in soils with time have been reported by many authors both for European cropping systems (Moolenaar and Lexmond, 1998; Keller et al. 2001, Keller and Schulin, 2003) as well as those in Canada (Sheppard et al., 2009), Australia (de Vries and McLaughlin, 2013) and China (Wang et al., 2014). The majority of published balances are limited to specific farms or farming systems and a spatially explicit analysis at the European level is still lacking. For the Netherlands a spatially explicit model was developed by Tiktak et al. (1998) which was able to explain regional trends in the Cd levels in soil; results indicated that in most cropping systems Cd levels in soil increased from 0.09 in 1930 to 0.27 mg kg^{-1} in 1990. Trends of increasing Cd levels in soils were observed in Australia as well (De Vries and McLaughlin, 2013) which basically suggests that in a large number of cropping systems positive Cd balances have dominated. Recent studies by Six and Smolders (2014) and Smolders (2017) for the EU, however seem to suggest that the Cd balance is, on average negative (average: -1.0 g Cd ha^{-1} yr^{-1}) which would lead to a decrease in the soil Cd content with time. The main reason for the deviation in the results by Six and Smolders (2014) and Smolders (2017) compared to other balance studies is the markedly higher calculated leaching loss which results in a net removal of Cd from soil in current cropping systems included in the assessment (potato and wheat). A second reason that has affected the Cd balance is the marked decrease in atmospheric deposition with time. For the Netherlands it was estimated that average deposition levels increased up to 2 g Cd ha^{-1} yr^{-1} until the mid-1980's but decreased sharply afterwards to values below 1 g Cd ha^{-1} yr^{-1}, present values being close to 0.58 g Cd ha^{-1} yr^{-1} as used in this study.

Data in table 4 indicate that the contribution from mineral fertilisers in arable land is the dominant source of Cd and contributes to 45% of the total load to soils for arable land and pasture combined. Atmospheric deposition however is still an important source of Cd inputs to agricultural soils despite the reduction that was achieved during the last decades. In grassland soils, inputs via atmospheric deposition is still larger than that of fertilisers. The contribution of biosolids at EU level is limited and amounts to only 4% of the total load to agricultural soils. Since it was assumed in the model that biosolids are applied only to arable soils, inputs to grassland are equal to zero. One of the current issues in the Integrator model is related to the fact that plot specific application rates of biosolids are lacking and an equal distribution among all arable land was assumed. Clearly biosolids are not applied equally across all land and the impact on the local Cd balance can be expected to be larger in those fields actually receiving bioslids compared to those not receiving biosolids. An overview by Nicholson et al. (2006) indicates that Cd loads from biosolid treated fields can be as high as 19 g Cd ha^{-1} yr^{-1} assuming that biosolids are used as prima source for N fertilisation. Clearly such application rates are not common but such data indicate that the local application of biosolids can significantly alter the (local) Cd balance. The contribution of animal manure to the total load of Cd is also limited (18%) which is due to the low levels of Cd in animal feed and additives commonly used in agriculture. At EU level, the major output of Cd from the soil occurs via leaching. Both in pasture soils and arable soils the net outflow via leaching exceeds inputs via fertiliser or atmospheric deposition. Crop uptake also leads to a net removal of Cd from soil but the average removal rate of 0.26 g Cd ha^{-1} yr^{-1} is 2 to 5 times lower than net leaching losses. Due to the lower soil pH and, on average, higher net water flows in pasture soils, the leaching from pasture is markedly higher (-1.21 g Cd ha^{-1} yr^{-1}) than that in arable soils (-0.55 g Cd ha^{-1} yr^{-1}) which is also the main reason why current Cd balances in pasture soils are negative compared to the slightly positive results in arable soils.

Table 4. Overview of Cd Inputs, Outputs and resulting balance at EU level in g Cd ha^{-1} yr^{-1} (left) and total load (in ton yr^{-1}) and the relative contribution of all fluxes (between brackets in) in 2017 for the Business as Usual scenario

	Cd Load (g Cd ha^{-1} yr^{-1})				Total load (ton Cd yr^{-1})		
	Grassland	Arable	Total		Grassland	Arable	Total
Surface Area	3.82E+07[1]	1.13E+08	1.52E+08		3.82E+07[1]	1.13E+08	1.52E+08
					ha	ha	ha
Manure	0.16	0.26	0.23		6.1	29.5	34.9 (18%)
Min. Fert.[2]	0.39	0.64	0.58		14.9	72.6	87.9 (45%)
Compost	0	0.02	0.01		0.0	2.3	1.5 (1%)
Sludge	0	0.06	0.04		0.0	6.8	6.1 (3%)
Atm. Dep.	0.43	0.42	0.42		16.4	47.6	63.7 (33%)
Plant Uptake	-0.26	-0.26	-0.26		-9.9	-29.5	-39.4 (27%)
Leaching	-1.21	-0.55	-0.71		-46.3	-62.4	-107.7 (73%)
Accumulation	-0.49	+0.59	+0.32		-18.7	+66.9	+48.5

[1] excluding rough grazing (non-managed grassland)

[2] including liming materials

One of the clear advantages of a spatially explicit approach is the capacity to perform an upscaling of the results from the 1x1 km grid level to NCU, country or even EU level (as presented in table 4). To illustrate the difference in inputs at country level the relative contribution of atmospheric deposition, manure, mineral fertiliser and manure for pasture soils and arable soils is given in figure 3. Striking difference can be observed for the contribution of atmospheric deposition which is still the dominant source of Cd in a.o. Bulgaria, Hungary, Romania and Slovakia for both pasture soils and arable soils. Relatively large contributions from animal manure are observed in countries with intensive livestock breeding systems like the Netherlands and Belgium. Inputs from mineral fertilisers are especially high in Spain and Portugal.

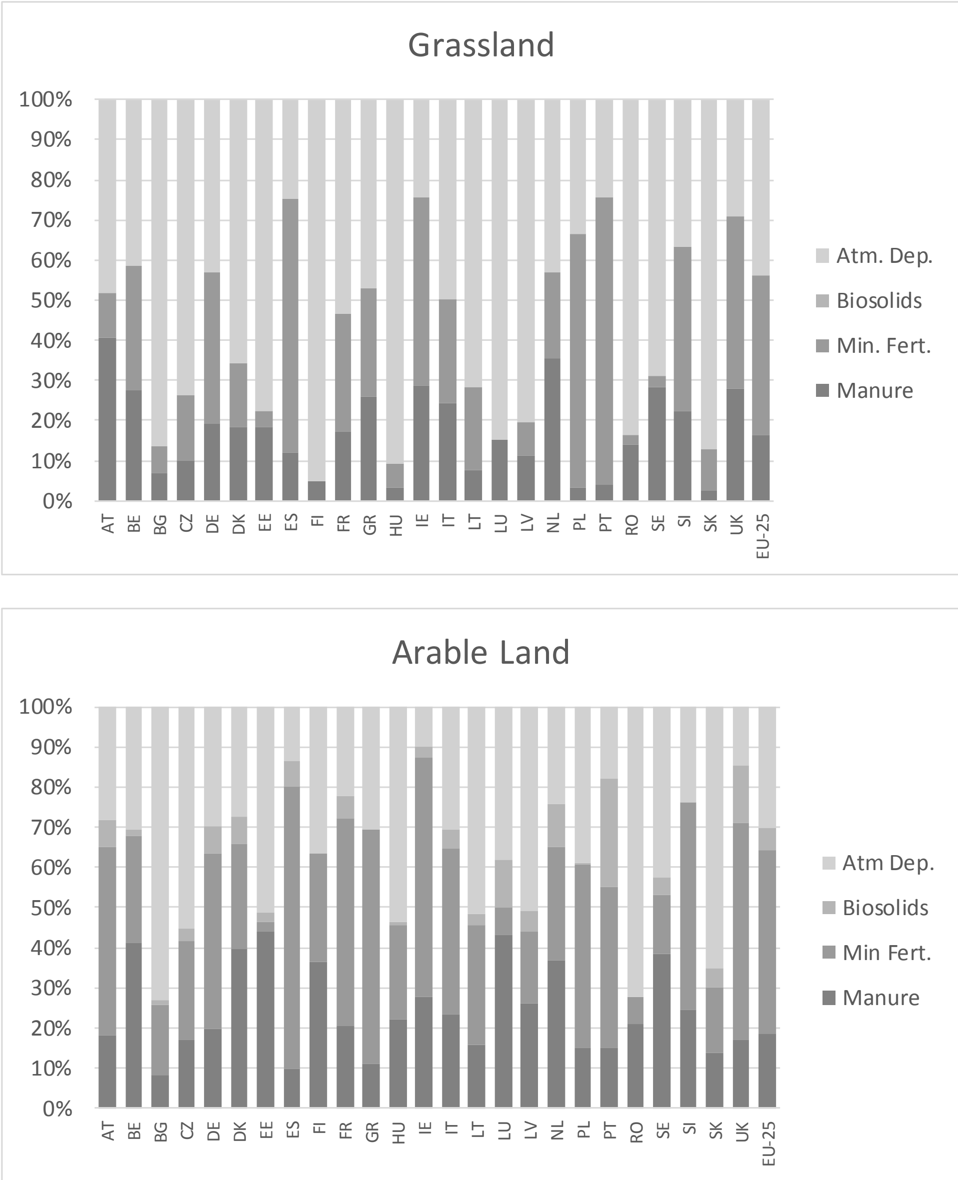
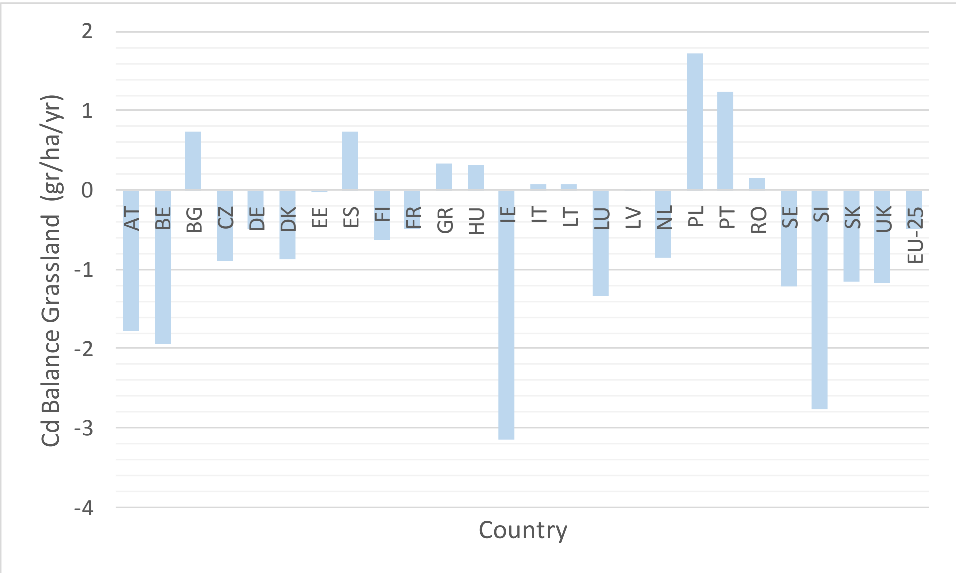


Figure 3. Variation in contribution of inputs at country level

In figure 4 the total balance (inputs – outputs) at country level is shown for pasture (top) and arable soils (bottom). For pasture soils, net depletion is commonly observed with quite explicit negative balances for a.o. Ireland, Slovakia, Belgium and Austria. In most cases this is related to a combination of a net high water leaching rate (high rainfall, limited evaporation) in combination with predominantly low pH soils which result in high predicted dissolved Cd concentrations in these countries. The positive balance for Poland, Portugal and Spain on the other hand is due to a high application rate of fertiliser (0.7 to 1.7 Cd ha^{-1} yr^{-1}), or atmospheric deposition (Bulgaria). For arable soils the distribution is similar to pasture soils albeit that most balances are shifted to a positive value. Exceptions for this trend are Belgium and the Netherlands which have a markedly more negative balance. This is largely due to the fact that in both countries the use of mineral P fertiliser is lower compared to other countries since many farmers use animal manure as the main source for P fertilisation and little mineral P fertiliser is required to match the crop demand. Since Cd levels in animal manure are lower than those in mineral fertiliser the Cd load to arable soils related to P fertilisation is low. Additional factors are a rather crop production rate (see figure 5) and high leaching losses due to both lower pH values (compared to calcareous soils) and higher water fluxes (compared to those in southern EU countries).



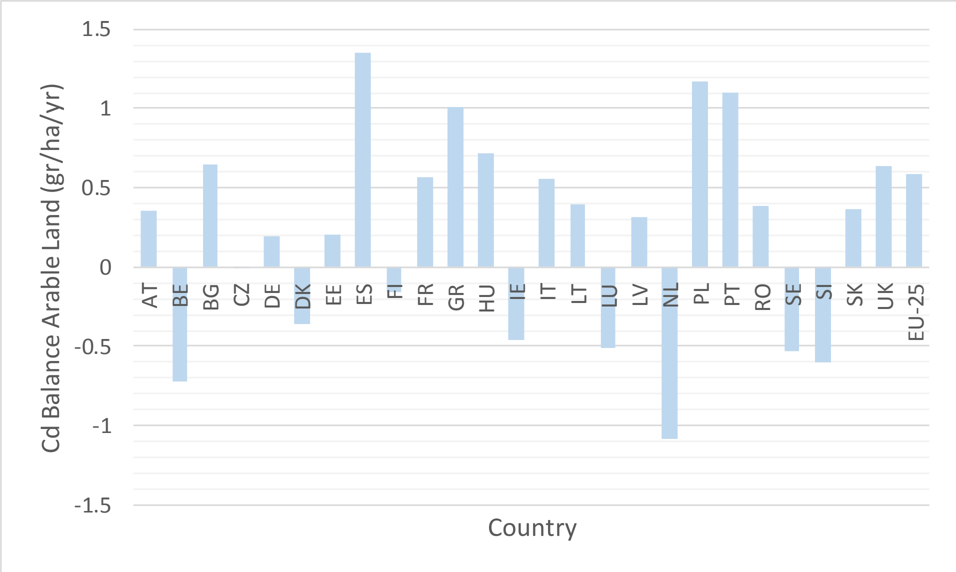


Figure 4. Overview of current Cd balance at country and EU-27 level in Grassland (top) and Arable soils (bottom), note the differences in the scale of the Y-axes.

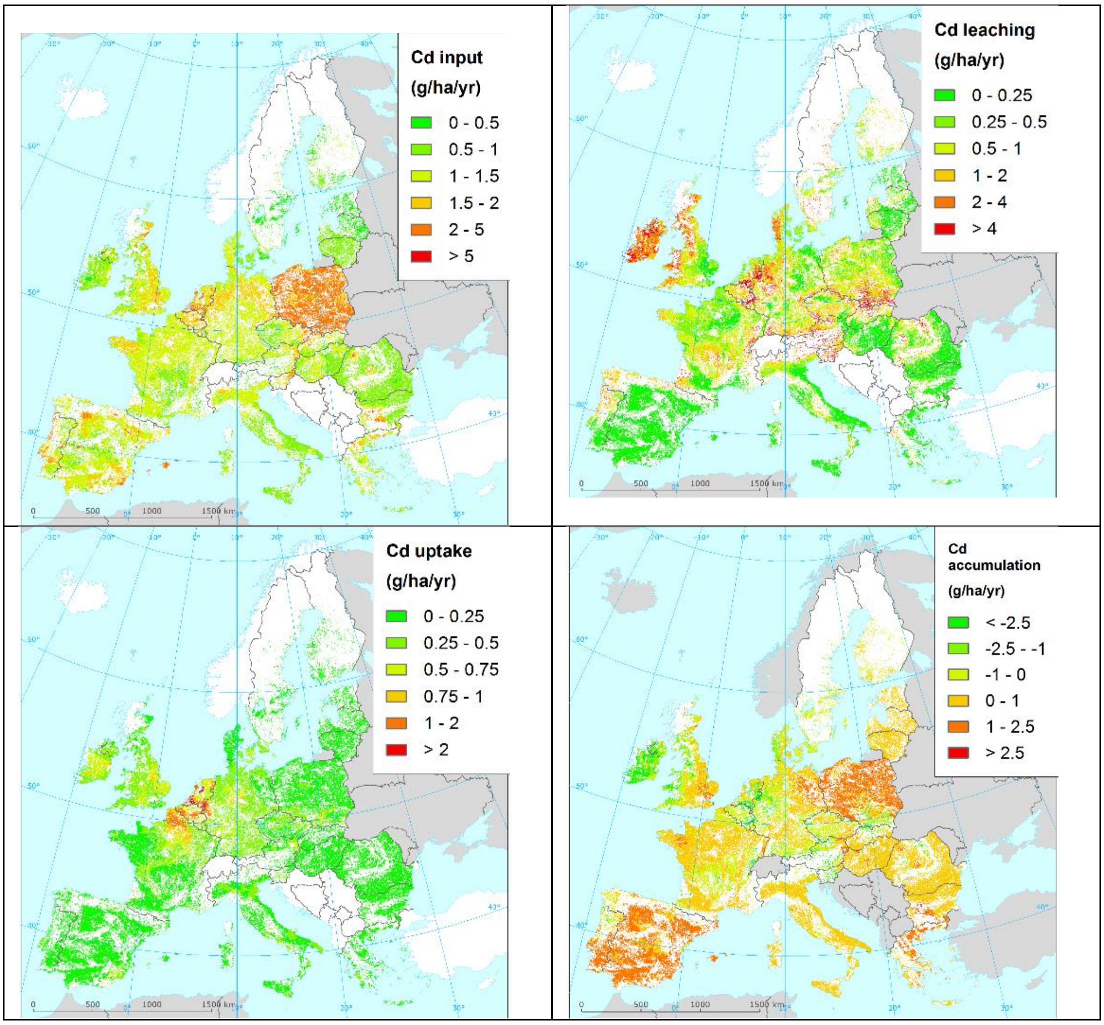


Figure 5 Spatial distribution of inputs, outputs and net Cd balance at EU level (data NCU)

Figure 5 presents the spatial distribution of the main Cd fluxes with a distinction between the two major outputs leaching and crop uptake. In general outputs from leaching exceed those via crop uptake and the graph with leaching losses clearly defines those areas with either high rainfall (NW Europe), low pH soils (central EU, UK, NW part of Portugal) and/or elevated Cd levels in soil (areas in Central Europe). The resulting map of Cd balances therefore shows a marked distribution across Europe which reflects a combination of both impact of soil properties or land use. The combination of high rainfall, low pH soils leads to negative balances in a.o. Ireland, Belgium and the Netherlands whereas accumulation is dominant in most of the Mediterranean countries as well as Central and Eastern Europe due to either the presence of high pH, fine textured soils in combination (for Spain and Poland especially) with high consumption rates of mineral P fertilisers and/or low leaching rates. At country level such differences lead to Cd balances (pasture and arable combined) that range from -2.5 g Cd ha^{-1} yr^{-1} for Ireland to +1.28 g Cd ha^{-1} yr^{-1} for Poland.

4. Impact of policy changes on Cd balance and Cd levels in soil

One of the aims of this paper was to assess to what extent changes in the allowed Cd content in fertiliser will affect the balance at regional (NCU/NUTS), national and EU level. To this purpose changes in the current balance (table 5, figure 5) at country level as well as long term changes in the soil Cd content (figure 6) have been calculated using the same model settings except for the allowed Cd content in mineral P fertilisers which varies between 0 and 80 mg Cd kg^{-1} P_2O_5 which represents the current proposed ranges (corresponding to a proposed maximum level of 60 mg Cd kg^{-1} P_2O_5 which is to be reduced to 40 and 20 mg Cd kg^{-1} P_2O_5 within a yet to be specified amount of years) as well as the zero input of Cd from mineral P fertilisers. Data in table 5 reveal that for pasture the balance for Cd will remain negative and only at the level of 80 mg Cd kg^{-1} P_2O_5 a net zero balance is observed. For arable land however, the reverse is true and stand-still, i.e. no accumulation is obtained only if no Cd is added to soil via mineral fertiliser (Cd-0 scenario). As was observed for the current balances and the variation therein, the impact of the proposed range in Cd levels in P fertilisers has difference consequences depending on the country. For arable land, which in view of food safety is perhaps of more relevance compared to pasture, balances in the Netherlands, Belgium, Sweden and Denmark remain largely negative and even at 60 mg Cd kg^{-1} P_2O_5 there is equilibrium between inputs and outputs. On the other hand, the majority of countries will not achieve a balance between inputs and outputs even when reducing Cd inputs from mineral fertiliser to zero. Apparently inputs from other sources than fertilisers are such that accumulation will continue even without inputs from fertilisers. IN figure 5 a spatial distribution of current balances (upper left) and changes in the balances relative to the current situation for the remaining scenarios (Cd-0 to Cd-80) is given. Clearly a reduction to zero inputs will lead to a reduction in the supply and hence a decrease of the accumulation as is reflected by the Cd-0 scenario map. The Cd-40 map clearly reveals the difference between countries which, at present, use below and above average Cd-P fertilisers. Where most countries at present use fertilisers with an average Cd content below or close to 40 mg Cd kg^{-1} P_2O_5, Poland, Spain and Portugal use fertiliser with, on average higher levels of Cd in P fertiliser which results in a reduced load compared to current loads if the limit were to be set at 40 mg Cd kg^{-1} P_2O_5. The Cd-60 and Cd-80 maps illustrate that at present the average Cd content used is below 60 (or 80) mg Cd kg^{-1} P_2O_5 which implies that these scenarios will lead to an increase in the accumulation as was shown at country level as well (table 5).

Table 5 Present surface weighted mean Cd balances at EU-27 (excl Malta and Cyprus) and country level (in g Cd ha^{-1} yr^{-1})

	Land Use											
	Grassland						Arable Land					
Country	BaU	Cd0	Cd20	Cd40	Cd60	Cd80	BaU	Cd0	Cd20	Cd40	Cd60	Cd80
EU-27	-0.49	-0.86	-0.64	-0.43	-0.21	0.00	0.59	0.00	0.37	0.75	1.12	1.49
AT	-1.78	-1.87	-1.82	-1.77	-1.72	-1.67	0.36	-0.33	0.05	0.44	0.82	1.20
BE	-1.93	-2.35	-1.97	-1.58	-1.20	-0.82	-0.72	-1.20	-0.76	-0.31	0.13	0.57
BG	0.74	0.67	0.76	0.85	0.94	1.02	0.65	0.49	0.70	0.91	1.12	1.33
CZ	-0.89	-1.04	-0.88	-0.72	-0.56	-0.40	-0.01	-0.26	0.00	0.27	0.54	0.80
DE	-0.49	-0.86	-0.57	-0.28	0.00	0.29	0.19	-0.28	0.09	0.46	0.83	1.20
DK	-0.87	-0.92	-0.87	-0.81	-0.75	-0.70	-0.36	-0.54	-0.36	-0.17	0.01	0.19
EE	-0.02	-0.03	0.11	0.25	0.38	0.52	0.20	0.20	0.37	0.55	0.72	0.89
ES	0.74	0.04	0.34	0.63	0.93	1.22	1.35	0.29	0.74	1.18	1.63	2.08
FI	-0.63	-0.63	-0.61	-0.60	-0.58	-0.57	-0.16	-0.16	0.08	0.32	0.57	0.81
FR	-0.50	-0.66	-0.56	-0.45	-0.34	-0.24	0.57	-0.11	0.33	0.78	1.23	1.67
GR	0.33	0.10	0.24	0.38	0.51	0.65	1.01	0.22	0.69	1.17	1.64	2.12
HU	0.31	0.27	0.31	0.35	0.39	0.42	0.72	0.48	0.70	0.92	1.14	1.36
IE	-3.14	-3.33	-3.21	-3.09	-2.97	-2.85	-0.46	-1.39	-0.81	-0.23	0.35	0.93
IT	0.07	-0.10	0.05	0.20	0.35	0.50	0.56	0.12	0.51	0.90	1.28	1.67
LT	0.07	-0.04	0.11	0.25	0.39	0.53	0.40	0.19	0.48	0.77	1.05	1.34
LU	-1.34	-1.34	-1.12	-0.91	-0.69	-0.48	-0.51	-0.51	-0.29	-0.08	0.14	0.36
LV	0.01	-0.02	0.03	0.08	0.13	0.18	0.32	0.21	0.41	0.61	0.80	1.00
NL	-0.86	-1.12	-0.97	-0.82	-0.67	-0.52	-1.08	-1.64	-1.32	-0.99	-0.67	-0.35
PL	1.72	-0.01	0.84	1.69	2.54	3.38	1.17	0.14	0.65	1.15	1.66	2.16
PT	1.25	0.15	0.53	0.91	1.28	1.66	1.10	0.38	0.63	0.88	1.12	1.37
RO	0.15	0.14	0.20	0.26	0.32	0.38	0.39	0.35	0.52	0.69	0.86	1.03
SE	-1.21	-1.21	-1.19	-1.17	-1.14	-1.12	-0.53	-0.54	-0.37	-0.20	-0.02	0.15
SI	-2.76	-3.34	-2.98	-2.62	-2.26	-1.90	-0.60	-1.40	-0.90	-0.40	0.10	0.61
SK	-1.15	-1.26	-1.12	-0.98	-0.84	-0.70	0.37	0.19	0.42	0.64	0.86	1.09
UK	-1.18	-1.47	-1.31	-1.16	-1.01	-0.85	0.64	-0.27	0.22	0.71	1.20	1.70

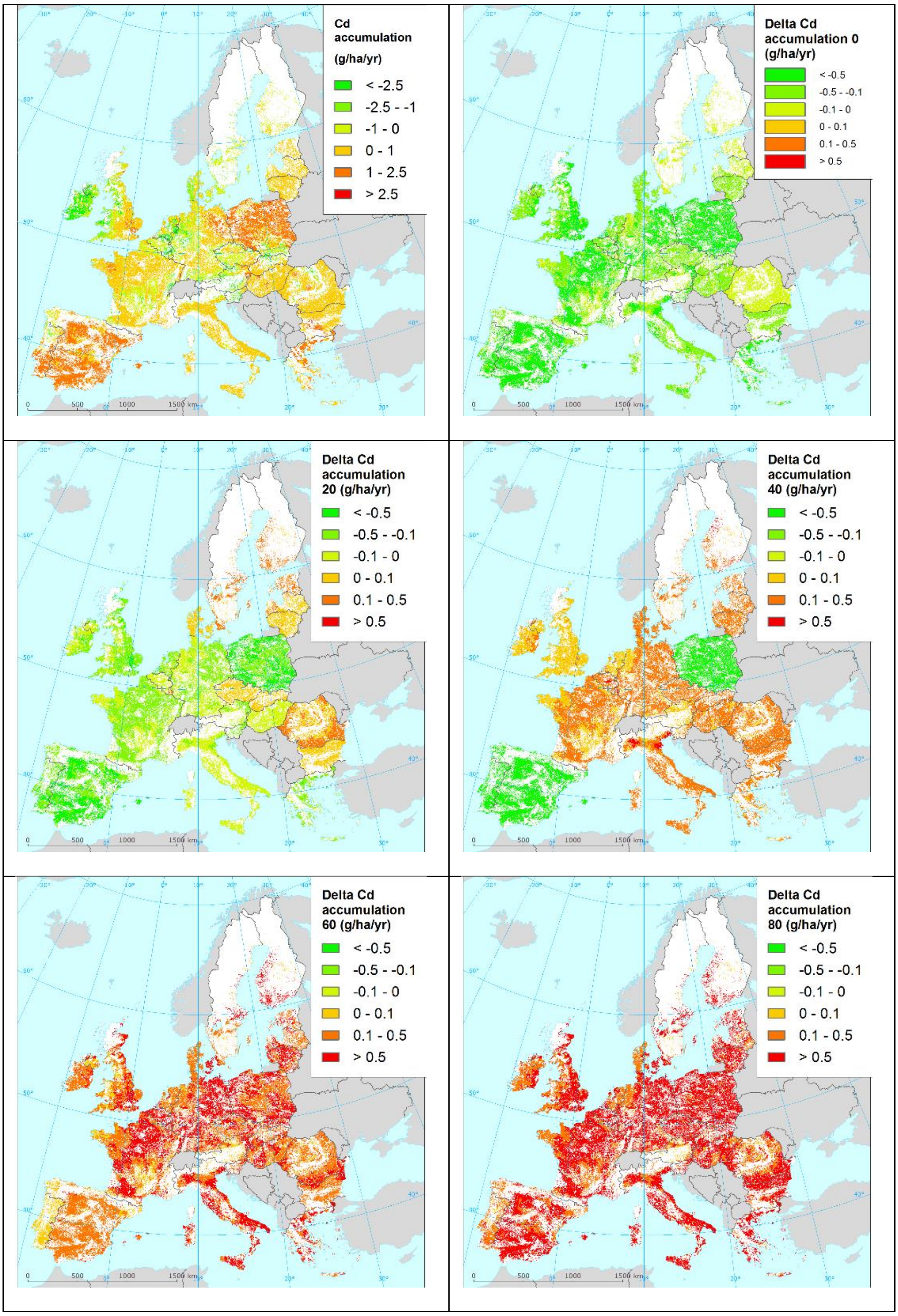


Figure 6. Current accumulation at EU level (top left) and changes in Cd balance (inputs – outputs) at EU level or the Cd-0 to Cd-80 scenario relative to the current situation.

Even though accumulation as such can be an indicator of the impact of proposed revision in the allowed cd content in P fertilisers, changes in the soil Cd ultimately are more important to address the consequences for the environment and transfer of Cd from soil to crops. In figure 6 and 7 changes in the Cd content at country level (figure 6) and across the EU (figure 7) are shown as predicted at t=100 years after the start of the simulation. At EU level changes in Cd levels in pasture soil are very limited and only in case of the Cd-80 scenario, average levels are predicted to increase with +3.8% compared to current Cd levels in soils (Table 7). And even though the changes in pasture soils in most countries are modest (-20 to +20% compared to current Cd levels in pasture soils), changes in the soil Cd content in Poland, Portugal and to a lesser extent Spain are substantial with Cd levels expected to double in both Portugal (increase from 0.09 to 0.2 mg kg^{-1}) and Poland (increase from 0.22 to 0.48 mg kg^{-1}).

For arable soils the shift from Cd-0 to Cd-80 largely results in an increase in the Cd content of the soil ranging from, at EU level, 0.2% in case of the Cd-0 scenario to +16% in case of the Cd-80 scenario (table 6). Table 6 also reveals that Cd levels in pasture soils are less prone to accumulation and an EU-wide increase in the soil Cd content is predicted to occur only in case of the Cd-80 scenario. When considering all agricultural land (with the exception of rough grazing areas including natural land), a zero net change in the Cd levels in soils is achieved at a level of approx. 20 mg Cd kg^{-1} P_2O_5 (table 6). At EU level current levels of Cd in mineral P fertilisers are such that soil Cd balances are close to equilibrium with a minor predicted increase (+2.4%) when considering the total area. For arable soils only however, balances are, as discussed positive and will lead to an, on average increase of +6.4% compared to current Cd levels in soils.

Table 6. Predicted relative change in the soil Cd content at EU-27 (excluding Malta and Cyprus) compared to current levels for grassland, arable and combined.

	Relative change in soil Cd levels at t=100 years from now		
Scenario	All Agricultural land	Arable	Grassland
BaU	2.4%	6.4%	-7.2%
Cd-0	-4.4%	0.2%	-15.6%
Cd-20	-0.1%	4.2%	-10.7%
Cd-40	4.1%	8.1%	-5.8%
Cd-60	8.3%	12.1%	-1.0%
Cd-80	12.5%	16.0%	3.8%

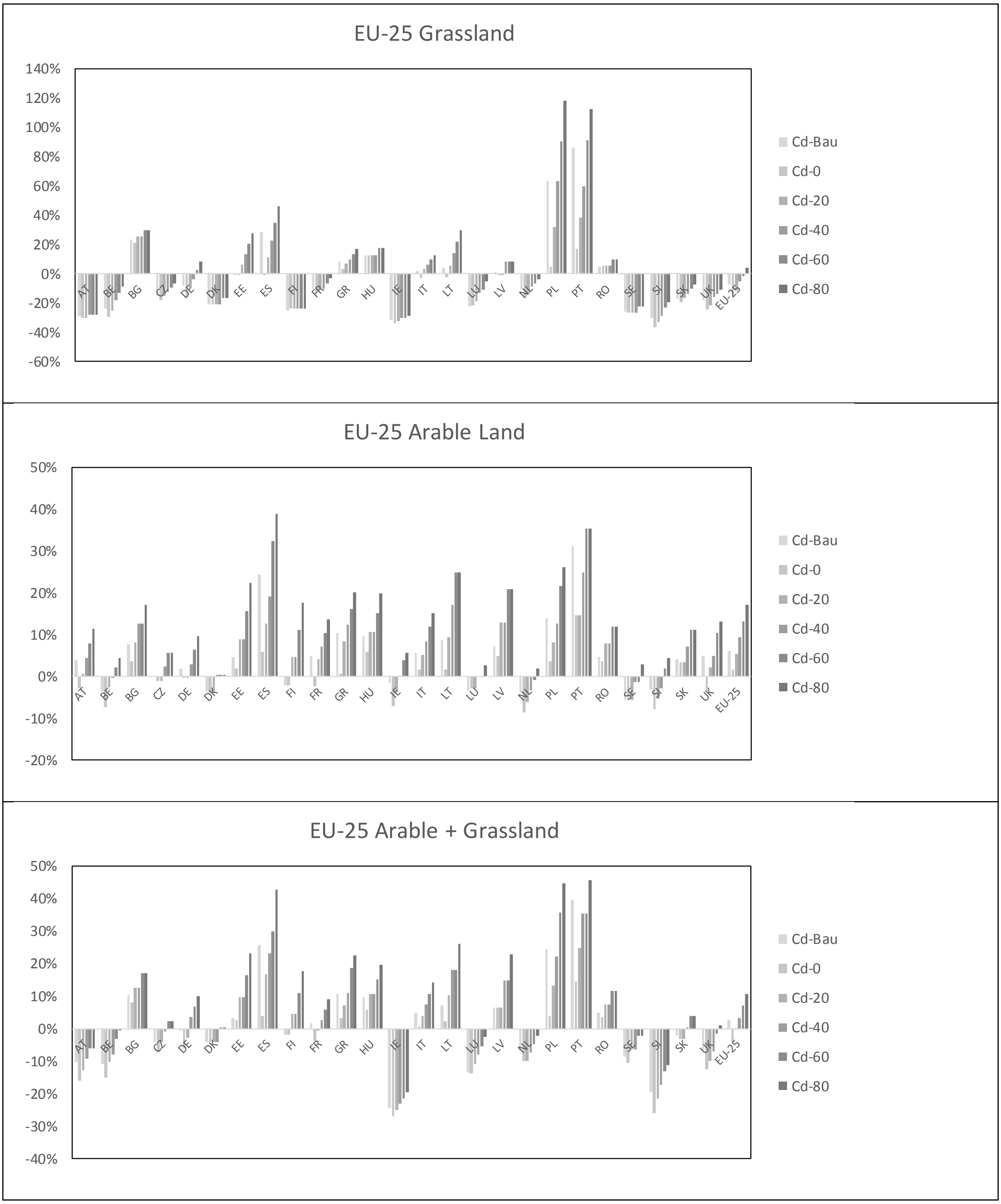


Figure 7. Average changes in soil Cd levels at country and EU level at t=100 years relative to current levels in soil for grassland (top), Arable land (middle) and all agricultural soils (bottom)

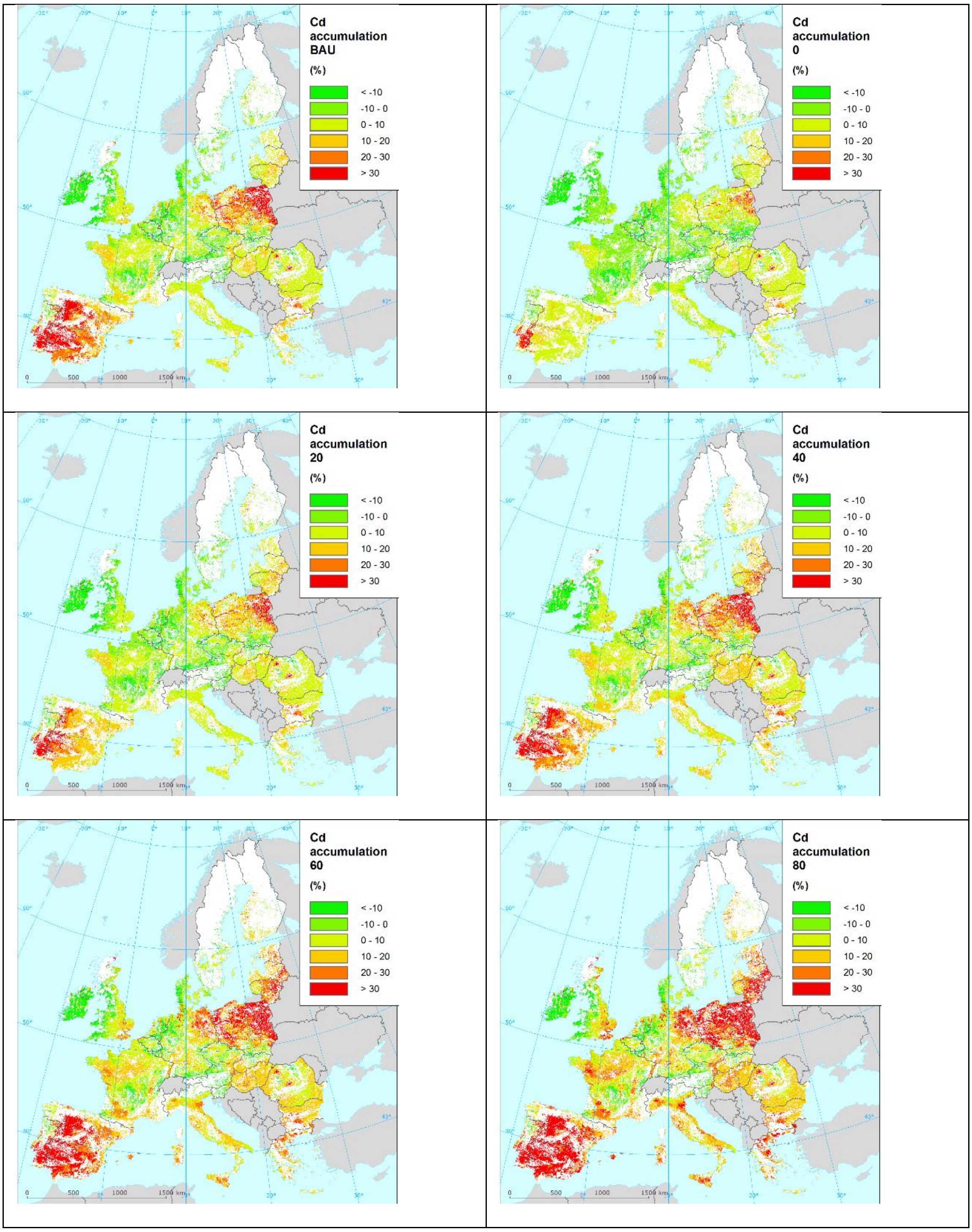


Figure 8. *Relative changes (%) in the soil Cd content after 100 years compared to 2010 (all soils, data at NCU level)*.

As illustrated in Figure 7, changes in soil Cd levels vary significantly between countries. At country levels the increase in the Cd levels in arable soils in a.o. Be, IE, Nl, Se and SL remains below 5% compared to current levels even in case of the Cd-80 scenario. This reflects a combination of a relatively low application of mineral P fertilisers (Nl), in combination with high predicted leaching losses from soil (Se, IE, Be, NL). On the other hand, Cd levels in soil but will increase substantially in countries like ES, PL and Pt regardless of the chosen level of Cd in mineral P fertilisers with predicted increases ranging from 14 to 30% in case of the Cd-0 scenario and between 35 to 38% in case of the Cd-80 scenario. These difference are related to both relatively high inputs via mineral P fertiliser on one hand (Pl) in combination with lower crop offtake rates and leaching (Es, Pt).

To illustrate how changes in the soil Cd content become more pronounced at a regional level, data in table 7 present the frequency distribution of the calculated changes in the soil Cd content at NUTS3 level; here both arable land and pasture is combined. The data in table 7 reveal that even in case of the Cd-0 scenario, the soil Cd content in more than 25% of the NUTS3 regions is expected to increase, where, on the other hand 25% of the regions also are close to equilibrium even in case of the Cd-80 scenario.

Table 7. Distribution of predicted changes in soil Cd at t=100 compared to t=0 at NUTS3 level

percentile	BaU	Cd-0	Cd-20	Cd-40	Cd-60	Cd-80
1	-35%	-38%	-36%	-35%	-33%	-32%
5	-24%	-28%	-25%	-23%	-21%	-19%
25	-5%	-9%	-6%	-3%	-1%	2%
50	2%	-3%	1%	4%	8%	11%
75	8%	2%	7%	11%	15%	20%
95	25%	10%	18%	27%	36%	45%
99	61%	24%	41%	58%	75%	91%
avg	+2%	-5%	-0%	+4%	+8%	+12%

Critical level of Cd in P fertilisers to achieve net zero balances in soil

An assessment was made of the critical Cd content in P fertilisers at which the Cd content in soil remains unchanged at t=100. This was done using data NUTS3 level for which changes in soil Cd were derived including both grassland and arable land. The underlying assumption was that changes in soil Cd were induced largely by a decrease in the Cd load from fertilisers; other inputs to soil remain constant (manure, biosolids, atmospheric deposition, other mineral fertilisers). Clearly changes in Cd content in soil are also affected by the changes in uptake and leaching but since changes in absolute levels of Cd in soil were relatively small, the impact of the changes in both outputs which are derived from the soil Cd content were also small. As a results, a highly significant positive linear relation was found between the imposed maximum levels of Cd in P fertiliser and the resulting relative changes of Cd in soil as is illustrated for a number of countries in figure 9 which presents the 5, 50 and 95 percentile values of the predicted changes in the soil Cd content at t=100 for each of the scenarios (Cd-0 to Cd-80)

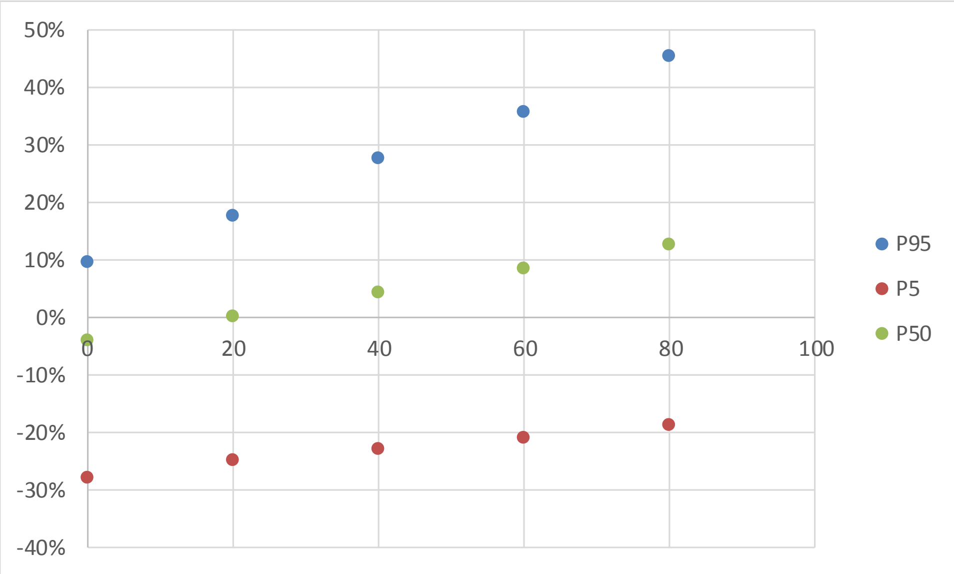


Figure 9. Five, fifty and ninety-five percentile of the predicted relative change of Cd in soil at the Cd-0, Cd-20, Cd-40, Cd-60 and Cd-80 scenarios at NUTS3 level for the combined arable and grassland soils.

Consequently the relationship between the predicted changes in soil Cd and the input concentration of Cd in fertiliser were used to back-calculate the critical Cd content in P fertilisers at which no change in soil cd would have been predicted. Based on the country averaged data (Table 6), the EU-wide critical level ranges from (a theoretical) -1.2 mg Cd kg^{-1} P_2O_5 for arable land to 64.2 mg Cd kg^{-1} P_2O_5 for grassland soils. For the total surface area covering both arable and grassland the mean critical level equal 20.7 mg Cd kg^{-1} P_2O_5 as shown in figure 9 as well. The negative value obtained for arable soils suggests that even if inputs via mineral P fertilisers were reduced to zero, inputs would still exceed outputs even though the balance is very close to zero.

5. Plausibility of results and impact of model choice on predicted changes in Cd levels in soil

One of the main findings as revealed by the Integrator model is that in order to obtain equilibrium between inputs and outputs, an EU-wide average level of 20 mg Cd kg^{-1} P_2O_5 would be required, whereas in fact a zero input from mineral P fertilisers would be required to reach stand still in arable soils. This result deviates substantially from those as obtained by Smolders (2017) who obtained a value of 73 mg

Cd kg^{-1} P_2O_5 as the critical value for arable soils (limited to wheat cropping and potato cropping systems only). From a conceptual model point of view the Smolders (2017) study is comparable to the one described here in that similar inputs and outputs are considered. In both approaches manure, mineral fertiliser, biosolids, atmospheric deposition and lime are considered as main inputs and also outputs, i.e. crop uptake and leaching from the topsoil are similar. In both the study by Smolders (2017) and this paper, inputs are largely based on existing data on quality of fertilisers, manure etc. and current application rates of such products. Consequently, calculated average levels of inputs to soil at EU level are in close agreement as shown in table 8. Since the model predictions from Smolders (2017) are derived for arable crops (potato and wheat) only, we use data from arable crops as calculated by Integrator for comparison purposes. Outputs from the soil in both cases are calculated using existing models. To calculate Cd removal via crop uptake, both models use a Bioconcentration factor assuming a linear transfer from soil to crop. Even though in the Integrator model more crops are considered compared to the ones in the study by Smolders (2017), the calculated average Cd removal rates by crops are also quite similar (table 8). Hence, the *partial* balance as calculated for the two models, which is also sometimes used in cases where leaching is not considered, is very comparable which confirms that despite differences in some of the assumptions used, predicted Cd balances are in the same order or magnitude.

Three major differences however can be identified between the approach presented in this paper (Integrator) and the one presented by Smolders (2017):

1. The model by Smolders is not based on spatial units but rather as a distribution of predictions from specific soil samples with known levels of Cd in soil and soil properties (GEMAS database, Reimann et a., 2014). Such an approach only would yield similar results if the database used by Smolders would be spatially representative for the entire surface covered by the database, which in its present form and data coverage is not the case (REF GEMAS).
2. Another difference is that Smolders (2017) only considered potato and wheat as relevant crops whereas the Integrator model is based on the true land use including 30 crops clustered in 11 specific crop groups, each with its own BCF. As discussed, this difference will not lead to major differences between the EU-wide average crop removal rate albeit that the data calculated for specific points are not necessarily representative for the actual removal rate due to differences in crops grown.
3. A large discrepancy between both approaches was observed for the leaching of Cd from the soil. EU-wide average leaching rates by Smolders (2017) are 2.0 g Cd ha^{-1} yr^{-1} (table 8) whereas this was 0.55 g Cd ha^{-1} yr^{-1} for arable soils using the Integrator model. The full balances, therefore,

which include leaching and is used to calculate changes of Cd levels in soils with time differ substantially with a net negative balance of -1.0 g Cd ha^{-1} yr^{-1} as calculated by Smolders (2017) compared to a net positive balance of +0.6 g Cd ha^{-1} yr^{-1} as calculated by Integrator. Even though such differences in the annual Cd load to soil are very small compared to the total Cd pool in soils which, on average, equals approximately 750 g Cd $ha^{-1}yr^{-1}$ (Smolders, 2017), such differences in the balance lead to markedly different predictions of the average relative change of Cd levels in soils for the scenarios evaluated in this paper as shown in table 9.

Table 8. Comparison of average inputs and outputs considered in the balance and resulting EU average Cd balance in current arable cropping systems (in g Cd $ha^{-1}yr^{-1}$; data at country level).

		Integrator (arable soils only)		
Input source	Smolders (2017)	Min EU-27	Average EU27	Max EU-27
Manure/biosolids/lime	0.2	0.09	0.34	1.10
Mineral fertiliser	0.7	0.01	0.64	1.08
Atmospheric deposition	0.3	0.16	0.42	0.91
Plant uptake	0.2	0.05	0.26	1.52
Partial balance (no leaching)	*+1.0*	*+0.30*	*+1.14*	*+2.15*
Leaching	2.0	0.08	0.55	2.51
Full Balance	*-1.0*	*-1.08*	*0.59*	*1.35*

Table 9 Predicted relative changes in soil Cd levels for the BaU and Cd-0 to Cd-80 scenario

	Relative change in soil Cd (in % compared to current Cd levels in soil)			
	Smolders (2017)	Integrator		
Scenario	EU-average	All soils	Arable	Grassland
Business as Usual	-16	2.4%	6.4%	-7.2%
Cd-0	Not included	-4.4%	0.2%	-15.6%
Cd-20	-21	-0.1%	4.2%	-10.7%
Cd-40	-13	4.1%	8.1%	-5.8%
Cd-60	-5	8.3%	12.1%	-1.0%
Cd-80	+3	12.5%	16.0%	3.8%

In both cases the water flux (in mm water percolating through the soil) used to derive the flux was comparable and the main difference in the outcome was, therefore, related solely to the model used to predict the Cd concentration in solution. Aside from the fact that the model used by Smolders (2017) to calculate Cd concentrations in the soil solution was based on different source data (n=151) and did not include a correction for the reactivity of Cd in soil as was done in case of the Integrator model, the main difference between the two models was the use of a linear model (n=1 in [eq]. 12) used by Smolders (2017) to calculate the Cd concentration in solution using pH and the soil organic matter content:

$$K_f = [Cd_{soil}]/[Cd_{solution}]^n \qquad [12]$$

With:

$$^{10}\log Kf = \text{constant} + \alpha_1 \cdot pH + \beta_1 \cdot \log(\text{Soil Organic Carbon}) \qquad [13]$$

With pH being pH $CaCl_2$ (0.01 M) as measured in soil extracts obtained by the GEMAS database. A crucial difference appears to be the assumed 1:1 linear relationship between Cd in soil and in solution as represented by [eq.] 12 using a value of 1 for n. In the current version of Integrator used in this study, the value of n was not a priori assumed to be 1 but instead a non-linear version of the Freundlich model ($n \neq 1$) was used as described by Römkens et al. (2004) and later on used by a.o. Groenenberg et al. (2012) for a number of metals. In this version the variation in the Cd concentration in solution is related directly to that of the soil Cd content and soil properties:

$$^{10}\log Cd_{solution} = \text{Constant} + \alpha_2 \cdot pH + \beta_2 \cdot \log(\text{Soil Organic Carbon}) + \gamma \cdot {}^{10}\log(\text{clay}) + \delta \cdot {}^{10}\log(Cd_{soil}) \qquad [14]$$

The critical difference between both approaches is the consideration of non-linearity in the model used in integrator as represented by the coefficient δ in equation [14] whereas the model used by Smolders implicitly assumes that the value of δ equals 1 which then would render the same model in both cases, that is, if the same soil properties are being used to explain the variation in Cd in solution. To assess the consequence of the impact of the choice of model, both models (eq. 13 and eq. 14) were derived from a similar set of source data (Römkens and Salomons, 1998) that contains measured data in field derived soil solution samples from arable soils and soils under pasture and forest. Here we only used solution pH and soil organic matter as variables to keep the model structure comparable as the one by Smolders (2017). In table 10 the ratio of the predicted Cd concentration in soil solution as calculated by both models is given for a selected number of combinations of pH and soil organic matter and relevant Cd ranges in soil (0.2 to 1.0 mg/kg).

Table 10. Ratio of calculated Cd concentrations in solution using the linear and non-linear models (eq. 13 and 14) based on the same source data (Römkens and Salomons, 1998). Common combinations of soil properties as present in EU soils are marked in grey.

Soil Properties used		Ratio of predicted Cd solution concentrations (Linear Kf/non-linear model)		
%SOM	pH	Cd_{soil} = 0.2	Cd_{soil} = 0.5	Cd_{soil} = 1.0
2	5	2.0	4.0	6.8
5	5	1.1	2.2	3.8
10	5	0.7	1.4	2.4
30	5	0.4	0.7	1.2
2	6	1.5	3.1	5.3
5	6	0.9	1.7	3.0
10	6	0.6	1.1	1.9
30	6	0.3	0.6	1.0
2	7	1.2	2.5	4.2
5	7	0.7	1.4	2.3
10	7	0.4	0.9	1.5
30	7	0.2	0.4	0.7

The data in table 10 clearly illustrate that in a substantial number of combinations of soil properties representative for a large area used for arable crop production, predicted Cd concentration in solution by the linear model is 1.2 to 6.8 times of those as obtained by the non-linear model. Only in soils with high (> 10%) organic matter predicted concentrations by the non-linear model are higher those obtained by the linear model. However, most of the organic soils are usually not used for arable crop production but are in use for grassland which was not considered in this comparison.

This significant difference in calculated values of Cd in solution was observed as well when using the source data underlying the models used by Smolders (2016), (de Greyse, pers. comm.) which suggests that the use of either a linear or non-linear relation between Cd in soil and Cd in solution results in significantly different estimates in the Cd concentration in solution, at least in the range of Cd in soils and the combinations of soil properties that prevail in arable soils. To further assess to what extent the choice of model would affect the model outcome, the linear Kf model used by Smolders was incorporated in the Integrator model to be used in the dynamic scenario analyses instead of the default model. This allowed us to calculate, at the most detailed level (NCU) the magnitude of the leaching fluxes. In table 11 a comparison is made between leaching fluxes at NCU level for both grassland and arable land using both models in Integrator both at time 0 and at t=100 years from now. Both models reveal the large range that exists in the calculated leaching fluxes related to different combinations of soil properties and climatic conditions. Extreme leaching rates with annual Cd removal rates of more than 5 g Cd ha^{-1} yr^{-1}

occur in both cases but are far more common (the predicted 75 percentile by the linear model equals 5.5 g Cd ha^{-1} yr^{-1} for arable soils and 7.4 g Cd ha^{-1} yr^{-1} in pasture soils). Also at NCU level, the median predicted leaching fluxes at t=0 calculated by the linear model are 5 to 6 times higher compared to those calculated by the non-linear model used in Integrator.

Table 11. Comparison of frequency distribution of predicted leaching losses at NCU level in Arable and Grassland soils (BaU scenario at t=0 and t=100) using the standard non-linear leaching model (INT) and the linear model (INT_S)

	Leaching flux Arable soils (g ha^{-1} yr^{-1})				Leaching flux Grassland soils (g ha^{-1} yr^{-1})			
	INT	*INT_S*	INT	*INT_S*	INT	*INT_S*	INT	*INT_S*
Percentile	t=0	t=0	t=100	t=100	t=0	t=0	t=100	t=100
1	0.01	0.06	0.01	0.08	0.01	0.06	0.02	0.08
5	0.03	0.15	0.03	0.18	0.03	0.21	0.05	0.25
25	0.16	1.0	0.19	1.0	0.22	1.3	0.27	0.96
50	0.43	2.5	0.48	2.2	0.58	3.3	0.60	1.5
75	1.0	5.5	1.0	3.7	1.3	7.4	1.2	2.1
95	2.8	14.8	2.5	6.2	3.8	19.3	2.2	3.1
99	5.4	27.6	4.2	8.1	6.6	31.9	3.0	4.1

Even though measurements of leaching fluxes are difficult to obtain and often prone to methodically induced bias, existing data seem to suggest that the predicted range in leaching fluxes by the INTEGRATOR model are reasonable. Data from Gray et al. (2003) for example indicate that leaching of Cd ranged from 0.27 to 0.86 g Cd ha^{-1} yr^{-1} in agricultural soils from New Zealand. A large Cd balancing study for German arable soils by Schütze et al. (2003) reports Cd leaching losses between 0.1 and 1.2 g Cd ha^{-1} yr^{-1} with a median value of 0.28 g Cd ha^{-1} yr^{-1} based on measured concentrations of Cd in leachates (Bielert, 1999). In the Canadian study by Sheppard et al. (2009) a Kd value of 1300 was used which corresponds to an annual leaching loss ranging from 0.1 to 1.2 g Cd ha^{-1} yr^{-1} (average 0.7 g Cd ha^{-1} yr^{-1}) in soils with a Cd content between 0.1 and 0.5 mg kg^{-1} and a net leaching rate of 300 mm $year^{-1}$. For Austria, Spiegel et al. (2003) used a non-linear model similar to the one applied in Integrator to calculate leaching losses based on work by McBride et al. (1997). Leaching losses for typical Austrian farming systems thus calculated range from 0.26 to 0.45 g Cd ha^{-1} yr^{-1} for arable cropping systems. An inventory of monitoring data in acid forest soils from Scandinavia (Sevel et al., 2009) on the other hand reveals that Cd leaching fluxes in forest soils range can be as high as 1 to 5 g Cd ha^{-1} yr^{-1}, values which are in line with the predicted leaching rates by Smolders et al (2017). However, in contrast to the other reported values the high fluxes reported by Sevel et al. (2009) are related to the low pH (3.5 – 4.5) of the forest soils included in the study which is approx. two units lower than pH values in arable cropping systems as used in this study and by Smolders (2017). The impact of pH on the leaching flux is illustrated in table 12 where average leaching fluxes (arable soils only) calculated at NCU level are clustered as a function of pH. Results from the acid soils (pH < 4.5) as calculated by the non-linear model agree well with the reported values by Sevel et al. (2009) whereas those calculated by the linear model appear extremely high. Due to the high leaching losses as calculated by both models, leaching rates at t=100 decreased substantially due to the reduction of Cd levels in soils with pH levels < 5.5. this effect is even more pronounced when using the linear Kd model compared to the non-linear model resulting in a 90% decrease of Cd in soil in those units with levels below 4.5.

Table 12. predicted changes in leaching rates (g Cd ha^{-1} yr^{-1}) and relative changes in Cd in soil (in %) at t=100 compared to t=0

	Integrator			Smolders		
pH class	Leaching t=0	Leaching t=100	ΔCd soil (%)	Leaching t=0	Leaching t=100	ΔCd soil (%)
< 4.5	4.7	2.6	-32%	43.7	4.4	-90%
4.5 – 5.5	1.9	1.7	0%	11.1	3.8	-50%
5.5 – 6.5	0.9	0.9	3%	5.0	3.3	-26%
6.5 – 7.5	0.3	0.3	8%	1.3	1.2	-2%
>7.5	0.02	0.02	20%	0.09	0.1	19%

Conclusions

- At EU level, 45% (88 ton yr^{-1}) of all inputs of Cd to agricultural soils is from mineral fertilisers followed by atmospheric deposition (33%, 64 ton yr^{-1}) and manure (18%, 35 ton yr^{-1}). At present, biosolids (compost and sludge) only contribute 4% (8 ton yr^{-1}) of the total inputs
- Removal of Cd from soil largely occurs through leaching (73%, equivalent to 108 ton yr^{-1}) and crop removal only contributes to 27% (39 ton yr^{-1})

- Present Cd balances in arable land are positive for arable land but negative for pasture soils, which is largely due elevated leaching losses that prevail in pasture soils due to the lower soil pH compare to arable soils.
- For arable soils, accumulation occurs at all proposed levels of Cd (Cd-20 to Cd-60), albeit that the predicted increase in Cd soil levels decreases from 12.1% (at EU level) in case of the Cd60 scenario to 4.2% in the Cd20 scenario (relative to current levels of Cd in soil)
- A stand-still level for Cd in arable soils at t=100 years from now is achieved only if the supply of Cd via mineral P fertiliser is reduced to zero. When considering the total surface area used for agriculture (i.e. arable land and pasture) a standstill is achieved at approx. 20 mg Cd kg^{-1} P_2O_5
- Regional variation in Cd balances is large with accumulation prevailing in the Mediterranean areas and Poland. This is due to either high (average) levels of Cd in mineral P fertilisers used (Pl, Pt) or a very low Cd removal rate from soil due to low water fluxes in most Mediterranean countries.
- Differences in the magnitude of the dissolved Cd concentration used to calculate the leaching flux is the main reason for the pronounced difference between model results presented by Smolders (2017) and those generated in the present study.
- The main reason for this is choice of model, it was shown here that a linear Kd model by definition leads to higher predicted leaching concentrations under the conditions that dominate EU soils (SOM, pH, Cd soil). Predicted leaching rates by the linear model as used by Smolders (2017) appear to be substantially higher than those previously reported.
- Differences between models operating at point level as used by Smolders (2017) and those based on predictions for the complete surface area (this study) are substantial which is largely due to the non-linear response of the leaching flux to the combination of soil properties. To obtain representative area-wide EU average values for Cd balances or changes in of Cd levels in soil, point based models therefore require a surface weighted representative distribution of all combinations of soil properties and Cd levels in soil which, at present is not the case.

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POssibly to be included

Balance at NUTS3 level

Arable	AM	FE	COM	SLU	DEP	UP	LE	BAL	AREA
min	0.00	0.01	0.00	0.00	0.08	0.01	0.01	-5.49	73
5%	0.05	0.08	0.00	0.00	0.19	0.06	0.07	-1.47	877
50%	0.28	0.57	0.03	0.05	0.39	0.28	0.62	0.40	45674
SW[1]:	0.26	0.64	0.02	0.06	0.42	0.26	0.55	0.59	
95%	0.94	1.37	0.06	0.24	0.91	1.04	2.79	1.46	353099
max	5.94	2.17	0.28	0.87	2.42	6.25	6.36	5.24	933601

Pasture	AM	FE	COM	SLU	DEP	UP	LE	BAL	Area
min	0.00	0.00	0.00	0.00	0.09	0.03	0.01	-4.84	20
5%	0.00	0.01	0.00	0.00	0.19	0.09	0.07	-2.36	363
50%	0.09	0.25	0.00	0.00	0.40	0.25	0.66	-0.14	12408
SW:	0.16	0.39	0.00	0.00	0.43	0.26	1.21	-0.49	-
max	0.95	2.65	0.00	0.00	2.53	0.86	5.91	3.04	561748
95%	0.49	1.32	0.00	0.00	0.93	0.64	3.04	1.22	119544

SW: Surface weighted EU average