Corrigendum

Corrigendum to ’Cadmium mass balance in French soils under annual crops: Scenarios for the next century’ [Science of the Total Environment 639 (2018) 1440–1452]

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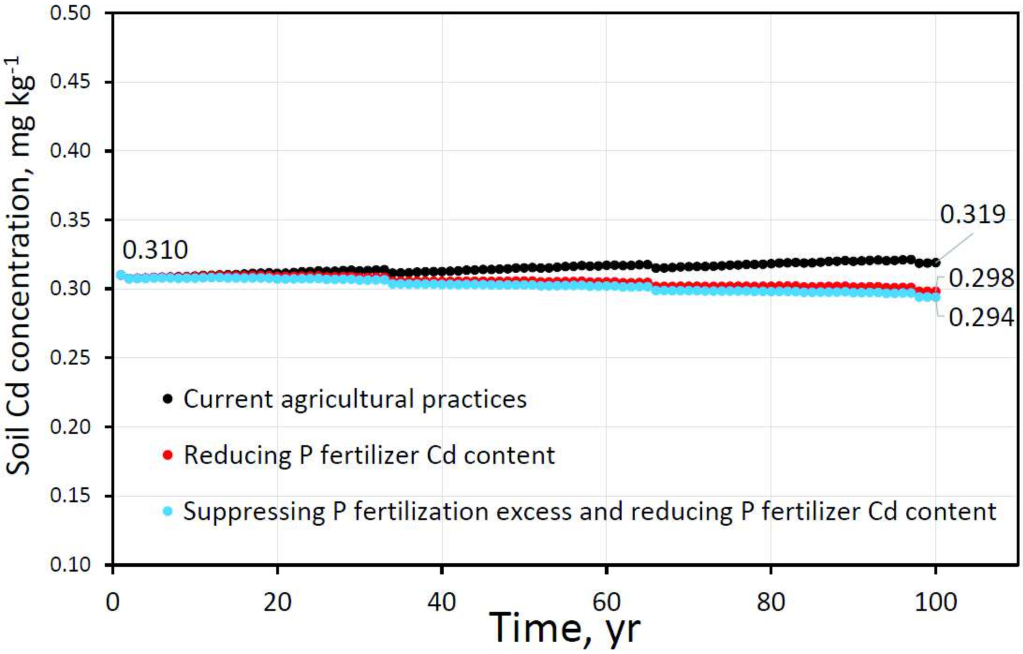
The authors regret an error in the data used for Cd input from inorganic P fertilizers. The mean current dose applied to each crop (AGRESTE, 2014) did not include the plots receiving no P fertilizer (*i.e.* the mean were calculated by AGRESTE excluding the plots receiving 0 kg P_2O_5 ha^{-1}). Therefore, the current mean P application rates were overestimated, which affected the results of the CPA and EUR scenarios. The other scenarios (GPPA, GPEU, OA and OAEU) were not affected by this error.
We calculated new P doses for each crop (including plots receiving no P fertilizer) and ran new simulations of the CPA and EUR scenarios. This led to a change in several tables, figures and sentences in the article and in the Supplementary Material.
The authors would like to apologize for any inconvenience caused.

The graphical abstract, Figure1, Figure 3 and Figure 4 should be as follows.



Graphical abstract

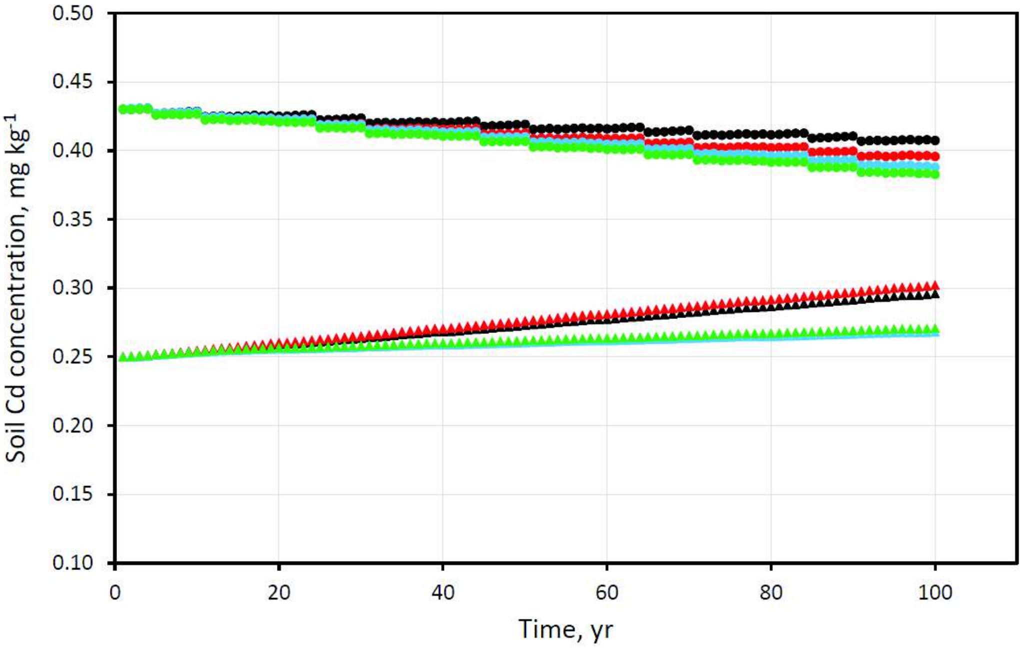


Fig. 1. Evolution of Cd concentration in the ploughed layer (25 cm) of soils under annual crops in the Centre (triangles) and Nord-Pas de Calais (circles) regions according to four scenarios with the lowest Cd leaching (Eq. (11)). Scenarios are CPA_{L12} (black), $GPPA_{L12}$ (red), EUR_{L12} (blue) and $GPEU_{L12}$ (green). Discontinuities in the Nord-Pas de Calais curves are due to the sugar beet offtake, resulting from a high yield (90.5 t ha^{-1}) and a relatively high TF (0.41). (For interpretation of the references to color in this figure legend, the reader is referred to the webversion of this article.)

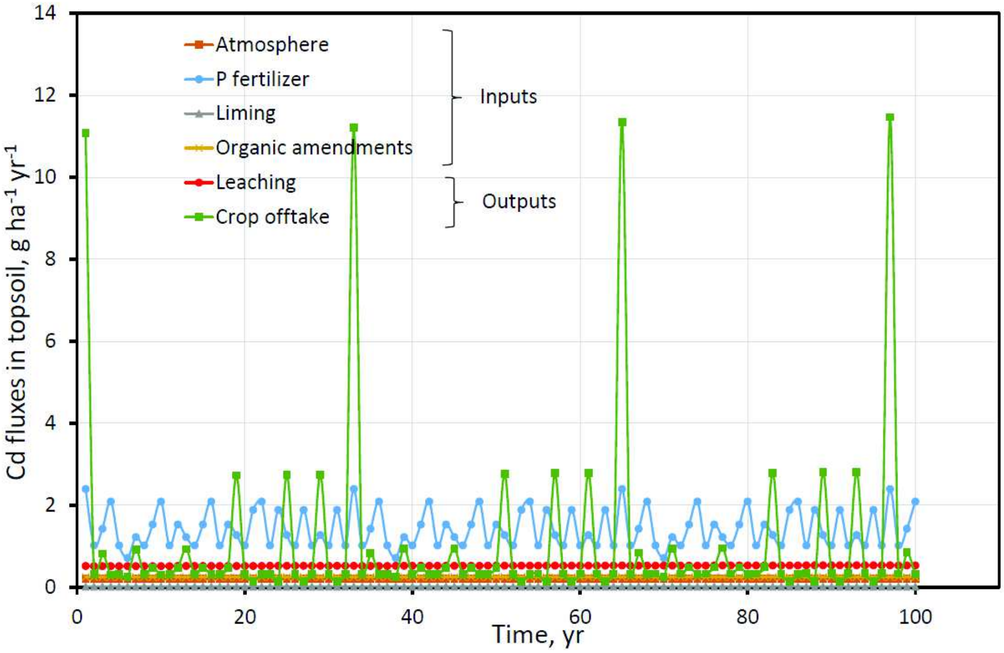


Fig. 3. Estimated annual cadmium fluxes in the ploughed layer of the mean French soil under annual crops for scenario CPAL$_6$.

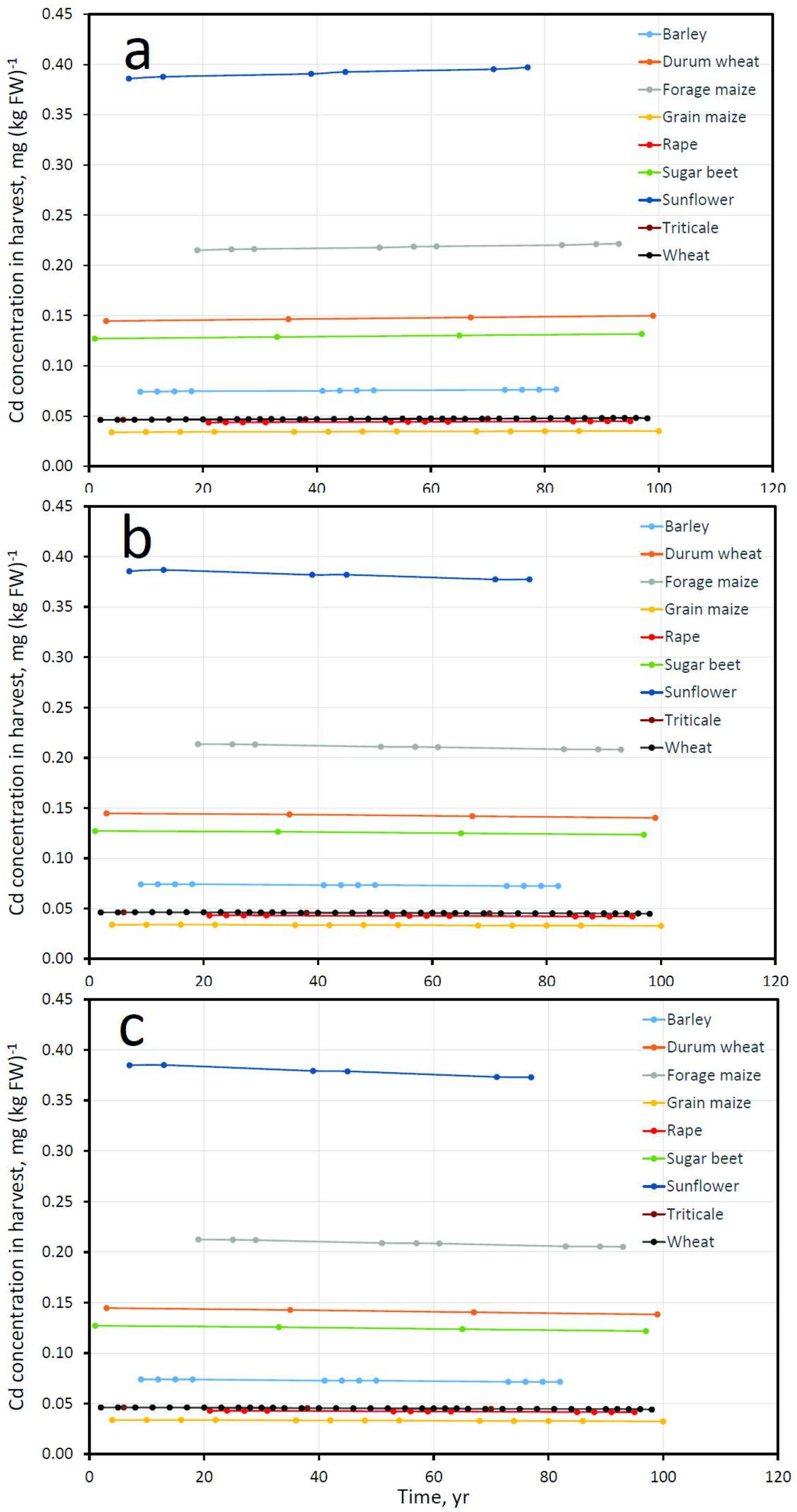


Fig. 4. Evolution of the estimated Cd content in crop harvests for three scenarios. a: $CPAL_6$ Scenario. b: $EURL_6$ Scenario. c: $GPEUL_6$ Scenario.

Tables 3, 4, 5, 6 and 7 should be as follows.

Table 1. Number of occurrences of each crop in the rotations, for each region and for France, for conventional agriculture. No data means no occurrence. For organic agriculture, eight field pea crops were added to the rotation simulated at national level.

Spatial unit	**Barley**	**Durum wheat**	**Forage maize**	**Grain maize**	**Potato**	**Rape**	**Sugar beet**	**Sunflower**	**Triticale**	**Wheat**
Alsace			1	11			1			4
Aquitaine			1	5				1		1
Auvergne	1		2	2		1		1	2	6
Bourgogne	6		1	1		6		1	1	10
Bretagne	1		6	2		1			1	6
Centre	3	1		2		4		1		8
Champagne-Ardenne	6		1	1		4	2			9
Corse	3		2	9					2	1
Franche-Comté	4		2	4		4		1	1	9
Ile de France	2			1		2	1			6
Languedoc-Roussillon	2	12				1		5	1	1
Limousin	2		5	1					5	4
Lorraine	2		1			2				3
Midi-Pyrénées	2	2	1	3		1		5	1	5
Nord-Pas de Calais	2		3		2	1	2			10
Basse-Normandie	1		5			1				6
Haute-Normandie	2		3		1	4	1			13
Pays de Loire	1		7	3		1		1	1	9
Picardie	2		1	1	1	3	3			12
Poitou-Charentes	2	1	1	4		2		4	1	9
Provence-Alpes-Côte d'Azur	3	13		1		1		2	1	2
Rhône-Alpes	2		3	6		1		1	1	6
France	*4*	*1*	*3*	*4*		*4*	*1*	*2*	*1*	*12*

Table 2. Evolution of Cd content in the ploughed layer (25 cm) of the French average soil under annual crops, according to different scenarios.

Scenario	Soil Cd content, mg kg^{-1}				Variation, %		
	Initial	10 yrs	20 yrs	100 yrs	10 yrs	20 yrs	100 yrs
CPA: Current P application rates	0.31	0.303	0.299	0.257	-2.2	-3.6	-17.3
CPA$_{L6}$: Same as CPA, with leaching rate /6	0.31	0.309	0.311	0.319	-0.3	0.4	2.9
CPA$_{L12}$: Same as CPA, with leaching rate /12	0.31	0.310	0.313	0.326	-0.2	1.0	5.2
GPPA: P application according to good practices	0.31	0.302	0.295	0.249	-2.7	-4.8	-19.7
GPPA$_{L6}$: Same as GPPA, with leaching rate /6	0.31	0.308[a]	0.309	0.310	-0.6	-0.5	-0.1
GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.31	0.309[a]	0.310	0.317	-0.4	0.0	2.2
EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.31	0.302	0.295	0.238	-2.5	-4.8	-23.2
EUR$_{L6}$: Same as EUR, with leaching rate /6	0.31	0.309	0.309	0.298	-0.4	-0.4	-3.8
EUR$_{L12}$: Same as EUR, with leaching rate /12	0.31	0.309	0.311	0.305	-0.3	0.3	-1.6
GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.31	0.301	0.294	0.234	-2.8	-5.3	-24.4
GPEU$_{L6}$: Same as GPEU, with leaching rate /6	0.31	0.308	0.307	0.294	-0.7	-0.9	-5.2
GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.31	0.308	0.308	0.301	-0.5	-0.5	-3.0
OA: Organic agriculture	0.31	0.302	0.296	0.248	-2.6	-4.7	-20.0
OA$_{L6}$: Same as OA, with leaching rate /6	0.31	0.308 [a]	0.309	0.310	-0.5	-0.3	0.0
OA$_{L12}$: Same as OA, with leaching rate /12	0.31	0.309 [a]	0.311	0.317	-0.3	0.2	2.3
OAEU: Same as OA, with EU regulation limiting Cd in fertilizers	0.31	0.302	0.294	0.237	-2.7	-5.0	-23.4
OAEU$_{L6}$: Same as OAEU, with leaching rate /6	0.31	0.308	0.308	0.298	-0.6	-0.6	-3.7
OAEU$_{L12}$: Same as OAEU, with leaching rate /12	0.31	0.309	0.309	0.305	-0.4	-0.2	-1.5

[a] This minimum value is the consequence of the high crop offtake of sunflower in year 7, made visible by a scenario with little variation in Cd balance over the century.

Table 3. Amounts of P_2O_5 applied to annual crops in France and in its region according to current practices (CPA) scenarios and to good practice (GPPA) scenarios. For France, the calculation is made either from the average of the regions or from the national balance model.

Spatial unit	**CPA**	**GPPA**	**Variation**
	kg P_2O_5 ha^{-1} (100 yrs)$^{-1}$		**%**
Alsace	5492	3830	-30
Aquitaine	4787	2195	-54
Auvergne	2801	1439	-49
Bourgogne	3513	2245	-36
Bretagne	1920	2395	25
Centre	3557	3994	12
Champagne-Ardenne	4349	3851	-11
Corse	3271	4464	36
Franche-Comté	3736	1788	-52
Ile de France	2004	3173	58
Languedoc-Roussillon	2873	2828	-2
Limousin	1736	1075	-38
Lorraine	2887	1885	-35
Midi-Pyrénées	3325	1946	-41
Nord-Pas de Calais	2645	1763	-33
Basse-Normandie	2258	1180	-48
Haute-Normandie	2403	2797	16
Pays de Loire	1357	690	-49
Picardie	1757	4097	133
Poitou-Charentes	2202	2376	8
Provence-Alpes- Côte d’Azur	1864	2728	46
Rhône-Alpes	3015	2067	-31
France (mean of Regions)	2807	2458	-12
France (nat. balance model)	2788	2098	-25

Table 4. Estimated mean cadmium fluxes (g Cd ha^{-1} yr^{-1}) in the ploughed layer (25 cm) of the mean French soil under annual crops for different scenarios.

Scenario	**Inputs**					**Outputs**			**Balance**
	P fertilizers	**Organic amendments**	**Liming**	**Atmo sphere**	**Total**	**Leaching**	**Crop offtake**	**Total**	
CPA: Current P application rates	1.42	0.25	0.02	0.20	1.89	2.83	0.93	3.76	-1.86
CPA$_{L6}$: Idem CPA, with leaching rate /6	1.42	0.25	0.02	0.20	1.89	0.53	1.04	1.56	0.33
CPA$_{L12}$: Idem CPA, with leaching rate /12	1.42	0.25	0.02	0.20	1.89	0.28	1.11	1.39	0.50
GPPA: P application according to good practice	1.07	0.25	0.02	0.20	1.54	2.78	0.91	3.69	-2.15
GPPA$_{L6}$: Idem CPA, with leaching rate /6	1.07	0.25	0.02	0.20	1.54	0.52	1.02	1.54	0.00
GPPA$_{L12}$: Idem CPA, with leaching rate /12	1.07	0.25	0.02	0.20	1.54	0.26	1.03	1.29	0.25
EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.64	0.25	0.02	0.20	1.11	2.74	0.90	3.63	-2.53
EUR$_{L6}$: Idem CPA, with leaching rate /6	0.64	0.25	0.02	0.20	1.11	0.51	1.00	1.51	-0.41
EUR$_{L12}$: Idem CPA, with leaching rate /12	0.64	0.25	0.02	0.20	1.11	0.26	1.02	1.27	-0.17
GPEU: P application according to good practice with EU regulation limiting Cd in fertilizers	0.47	0.25	0.02	0.20	0.94	2.71	0.89	3.60	-2.66
GPEU$_{L6}$: Idem GPEU, with leaching rate /6	0.47	0.25	0.02	0.20	0.94	0.51	1.00	1.50	-0.56
GPEU$_{L12}$: Idem GPEU, with leaching rate /12	0.47	0.25	0.02	0.20	0.94	0.26	1.01	1.26	-0.32
OA: Organic agriculture	0.83	0.08	0.02	0.20	1.13	2.77	0.55	3.32	-2.19
OA$_{L6}$: Same as OA, with leaching rate /6	0.83	0.08	0.02	0.20	1.13	0.52	0.60	1.12	0.01
OA$_{L12}$: Same as OA, with leaching rate /12	0.83	0.08	0.02	0.20	1.13	0.26	0.61	0.87	0.26
OAEU: Same as OA, with EU regulation limiting Cd in fertilizers	0.40	0.08	0.02	0.20	0.70	2.72	0.54	3.26	-2.56
OAEU$_{L6}$: Same as OAEU, with leaching rate /6	0.40	0.08	0.02	0.20	0.70	0.51	0.59	1.10	-0.40
OAEU$_{L12}$: Same as OAEU, with leaching rate /12	0.40	0.08	0.02	0.20	0.70	0.26	0.60	0.86	-0.16
Belon et al (2012), France	1.02	0.56	0.00	0.25	1.83	n.d.	n.d.	n.d.	n.d.
Six and Smolders (2014), Europe	0.79	0.06	0.09	0.35	1.29	2.56	0.20	2.76	-1.47

Table 5. Mean cadmium input with P fertilizer application and Cd crop offtake for each crop and three contrasting scenarios.

Crop	P fertilizer input				Offtake		
	CPA_{L12}	$GPEU_{L12}$	$OAEU_{L12}$	CPA_{L12}-$GPEU_{L12}$	CPA_{L12}	$GPEU_{L12}$	$OAEU_{L12}$
	g Cd ha^{-1} yr^{-1}			% CPA_{L12}	g Cd ha^{-1} yr^{-1}		
Barley	1.53	0.33	0.23	78.5	0.49	0.47	0.35
Durum wheat	1.43	0.58	0.45	59.3	0.85	0.82	0.62
Field pea			0.49				0.11
Forage maize	1.28	0.52	0.38	59.5	2.81	2.69	2.03
Grain maize	2.09	0.62	0.48	70.1	0.32	0.31	0.23
Rape	1.89	0.69	0.51	63.6	0.15	0.14	0.11
Sugar beet	2.40	1.05	0.82	56.2	11.40	10.97	8.27
Sunflower	1.22	0.03	0.50	97.6	0.95	0.92	0.69
Triticale	0.71	0.22	0.16	69.3	0.26	0.25	0.19
Wheat	1.02	0.40	0.30	60.4	0.33	0.32	0.24

In the new version of the Supplementary Material, Tables S2, S13, S14, S15 and S16 were corrected.

The text should be as presented in Table C1.

Table C1. Corrections of the text in relation with the new simulations of the CPA and EUR scenarios.

Section	**Sub-section**	**Initial sentence (incorrect)**	**Corrected sentence**
Highlights		• In France, P fertilizer applications represent around 85% of the soil Cd inputs. • Maintaining current cultivation practices, soil Cd content would increase by ca 15%. • Lessening both P fertilizer application and Cd content would reduce soil Cd content.	• In France, P fertilizer applications represent around 74% of the soil Cd inputs. • Maintaining current cultivation practices, soil Cd content would increase by 3-5%. • Lessening both P fertilizer application and Cd content would reduce soil Cd content.
Abstract		If current cultivation practices are maintained, the average Cd content would increase by about 15% after a century, due to the input of Cd with P fertilizer applications.	If current cultivation practices are maintained, the average Cd content would increase by about 3% after a century, due to the input of Cd with P fertilizer applications.
		This represents around 85% of the soil Cd inputs and is nearly twice the Cd output caused by leaching and crop offtake.	This represents around 74% of the soil Cd inputs and is nearly equal to the Cd output caused by leaching and crop offtake.
		These results conflict with those recently obtained at the European level, due to three factors: the higher rate of P application in France than in Europe, a higher Cd content in the P fertilizers applied in France and a lower Cd leaching in French soils.	These results conflict with those recently obtained at the European level, due to two factors: a higher Cd content in the P fertilizers applied in France and a lower Cd leaching in French soils.
		Assuming the current excessive P fertilization, the enforcement of a regulation limiting Cd content in the P fertilizers, as proposed by the European Union, would lead to a lesser increase in soil Cd, by 1.6% to 3.9% after a century.	Assuming the current P fertilization, the enforcement of a regulation limiting Cd content in the P fertilizers, as proposed by the European Union, would lead to a decrease in soil Cd, by 1.6% to 3.8% after a century.
1. Introduction		The mean P application rates on wheat and potato crops in France are 53 kg P_2O_5 and 84 kg P_2O_5 respectively, while those used at European level are 21 P_2O_5 and 45 P_2O_5, respectively (Six and Smolders, 2014).	*The initial sentence should be deleted*
3. Results and discussion	3.1. Future trends in French soil Cd content	In the case of simulations with the highest leaching rate (Eq. (8)), if current P fertilization practices are maintained in the future, i.e. when considering the CPA scenarios, the soil Cd content should slightly decrease ($V_{Cd} = -6.3\%$) after 100 years (Table 4). Alternatively, if suddenly, farmers strictly adopted the best practice for P application, as	In the case of simulations with the highest leaching rate (Eq. (8)), if current P fertilization practices are maintained in the future, i.e. when considering the CPA scenarios, the soil Cd content should decrease by 17.1% after 100 years (Table 4). Alternatively, if suddenly, farmers strictly adopted the best practice for P application, as simulated in the

		simulated in the GPPA scenario, the P fertilizer rates would be half those of current application rates, leading to a reduction of 19.7% of the Cd content in soils after 100 years, again supposing Cd leaching at its highest level. Enacting the EU regulation limiting Cd content in P fertilizers (EUR scenario) would cause a decrease of 18.4% in the soil Cd content.	GPPA scenario, the P fertilizer rates would be 12% to 25% lower than those of current application rates (Table 5), leading to a reduction of 19.7% of the Cd content in soils after 100 years, again supposing Cd leaching at its highest level. Enacting the EU regulation limiting Cd content in P fertilizers (EUR scenario) would cause a decrease of 23.2% in the soil Cd content.
		However, simulations with 12 times lower leaching (Eq. (11)) gave very different predictions. When considering the CPA_{L12} scenario, the soil Cd content should increase by 17.4% after 100 years (Table 4). Scenarios GPPA_{L12} and EUR_{L12} predicted small increases in soil Cd content, of 2.2% and 3.9%, respectively.	However, simulations with 12 times lower leaching (Eq. (11)) gave very different predictions. When considering the CPA_{L12} scenario, the soil Cd content should increase by 5.2% after 100 years (Table 4). Scenario GPPA_{L12} predicted a small increase in soil Cd content (= 2.2%), while EUR_{L12} predicted small decrease in soil Cd content (= -1.6%).
		In the case of an intermediate Cd leaching rate (Eq. (12)), CPA_{L6} predicted an increase of 15.0% in soil Cd content. According to GPPA_{L6}, soil Cd content would not vary ($V_{Cd} = -0.1\%$) after 100 years and slightly increase according to EUR_{L6} (V_{Cd}=1.6%).	In the case of an intermediate Cd leaching rate (Eq. (12)), CPA_{L6} predicted a slight increase of 2.9% in soil Cd content. According to GPPA_{L6}, soil Cd content would not vary ($V_{Cd} = -0.1\%$) after 100 years and slightly decrease according to EUR_{L6} ($V_{Cd} = -3.8\%$).
		Bases on the fact that dividing the maximum leaching rate (Eq. (8)) by 6 or 12 give close predictions, CPA_{L6} would be more reliable than CPA in predicting the trend of soil Cd content in a future where “business as usual” would be the way to manage annual crops. In this case, topsoil Cd content would increase by around 15% after a century.	Bases on the fact that dividing the maximum leaching rate (Eq. (8)) by 6 or 12 give close predictions, CPA_{L6} would be more reliable than CPA in predicting the trend of soil Cd content in a future where “business as usual” would be the way to manage annual crops. In this case, topsoil Cd content would increase by around 3% after a century.
		Scenario GPPA_{L6} predicts no variation in soil Cd content over the next century. However, this scenario gives more an assessment of the weight of fertilization practices on the soil Cd content (by comparison with the CPA scenarios) than a realistic prediction. Indeed, it seems very unlikely that all the farmers would simultaneously and immediately reduce their P applications by around 50%, strictly applying the COMIFER (1995) recommendations.	Scenario GPPA_{L6} predicts no variation in soil Cd content over the next century. It seems unlikely that all the farmers would simultaneously and immediately reduce their P applications by around 25%, strictly applying the COMIFER (1995) recommendations.
		Moreover, if the application of mineral P fertilizer did indeed declined between 1989 and 2008, its usage has been steady since then (UNIFA, 2014).	However, it is likely that in one to two decades, excess phosphate fertilization could be significantly reduced, by strengthening advice to farmers.
		The EUR scenarios, which predict a slight increase in soil Cd content, appear more realistic, as the project of a regulation reducing Cd in P fertilizers has existed at least since 2003 (DG Enterprise, 2003) and	The EUR scenarios, which predict a slight decrease in soil Cd content, appear more realistic, as the project of a regulation reducing Cd in P fertilizers has existed at least since 2003 (DG Enterprise, 2003) and

		especially as the European Commission produced a regulation proposal in 2016 (European Commisison, 2016).	especially as the European Commission produced a regulation proposal in 2016 (European Commisison, 2016).
		Supposing the maximum increase in the P fertilizer price due to decadmiation of €100 (t P_2O_5)$^{-1}$, this would increase the cost of the P fertilization for wheat (53 kg P_2O_5 ha^{-1}) by €5.3. This cost increase represents approximately 0.3% of the wheat production cost (1640 € ha^{-1} in 2014, Carpentier (2014)).	Supposing the maximum increase in the P fertilizer price due to decadmiation of €100 (t P_2O_5)$^{-1}$, this would increase the cost of the P fertilization for wheat (20 kg P_2O_5 ha^{-1}) by €2.0. This cost increase represents approximately 0.1% of the wheat production cost (1640 € ha^{-1} in 2014, Carpentier (2014)).
		Scenarios OA and OAEU are also quite unrealistic, as the total and rapid conversion of French agriculture to organic agriculture practices is very unlikely.	Scenarios OA and OAEU are quite unrealistic, as the total and rapid conversion of French agriculture to organic agriculture practices is very unlikely.
		Increasing the time lapses of application of the threshold contents resulted in a higher increase (EUR_{L6}:+0.5%) or a lower decrease (GPEU_{L6}: −0.2%; OAEU_{L6}: −0.1%) of soil Cd. However, the differences in V_{Cd} between the two regulation variants were small.	Increasing the time lapses of application of the threshold contents resulted in a lower decrease (EUR_{L6}: −0.2%; GPEU_{L6}: −0.2%; OAEU_{L6}: −0.1%) of soil Cd. However, the differences in V_{Cd} between the two regulation variants were very small.
		In the Centre, scenario CPA_{L12} predicted an increase in the soil Cd content of 30.5% after a century, while in the Nord-Pas de Calais, it predicted only a 3.4% increase.	In the Centre, scenario CPA_{L12} predicted an increase in the soil Cd content of 18.4% after a century, while in the Nord-Pas de Calais, it predicted a 5.3% decrease.
3. Results and discussion	3.2. Cadmium fluxes	In the scenarios with current P application rates (CPA, CPA_{L6}, CPA_{L12}), the total inputs were 3.31 g Cd ha^{-1} yr^{-1}, P fertilizer application accounting for 2.84 g Cd ha^{-1} yr^{-1}, that is 85.5% of the Cd input in soil. Cadmium from atmospheric deposition accounted for 6.1% of the inputs, while the amendments each represented <2.0% of the Cd inputs (Table S16).	In the scenarios with current P application rates (CPA, CPA_{L6}, CPA_{L12}), the total inputs were 1.89 g Cd ha^{-1} yr^{-1}, P fertilizer application accounting for 1.42 g Cd ha^{-1} yr^{-1}, that is 73.8% of the Cd input in soil. Cadmium from atmospheric deposition accounted for 11.1% of the inputs, while the amendments each represented <4.0% of the Cd inputs (Table S16).
		For the CPA scenario, the total outputs were 3.99 g Cd ha^{-1} yr^{-1} with Cd leaching of 3.00 g Cd ha^{-1} yr^{-1} (83.4% of the outputs) and 0.99 g Cd ha^{-1} yr^{-1} as crop offtake. In CPA_{L6} and CPAL_{12}, leaching was 0.56 g Cd ha^{-1} yr^{-1} (53.8% of the outputs) and 0.28 g Cd ha^{-1} yr^{-1} (38.9% of the outputs), respectively. The leaching flux was between 2.7 and 3.0 g Cd Cd ha^{-1} yr^{-1} when calculated according to Eq. (8).	For the CPA scenario, the total outputs were 3.76 g Cd ha^{-1} yr^{-1} with Cd leaching of 2.83 g Cd ha^{-1} yr^{-1} (83.4% of the outputs) and 0.93 g Cd ha^{-1} yr^{-1} as crop offtake. In CPA_{L6} and CPAL_{12}, leaching was 0.53 g Cd ha^{-1} yr^{-1} (53.8% of the outputs) and 0.28 g Cd ha^{-1} yr^{-1} (38.9% of the outputs), respectively. The leaching flux was between 2.7 and 2.8 g Cd Cd ha^{-1} yr^{-1} when calculated according to Eq. (8).
		In conventional agriculture, a reduction in the Cd content in the P fertilizer (EUR scenario) would reduce the average Cd input of 55% compared to the CPA scenario (from 2.84 to 1.27 g Cd ha^{-1} yr^{-1}, Table 6). The application of good practices for P fertilization (GPPA scenario) would have a similar effect to that of the EUR scenario, with a Cd input due to P fertilizer of 1.07 g Cd ha^{-1} yr^{-1}.	In conventional agriculture, a reduction in the Cd content in the P fertilizer (EUR scenario) would reduce the average Cd input of 55% compared to the CPA scenario (from 1.42 to 0.64 g Cd ha^{-1} yr^{-1}, Table 6). The application of good practices for P fertilization (GPPA scenario) would have an effect half that of the EUR scenario, with a Cd input due to P fertilizer of 1.07 g Cd ha^{-1} yr^{-1}.

		Excluding the scenario with leaching according to Eq. (8), only scenarios GPEU and OAEU (L6 and L12) led to a negative balance, of the order of −0.2 to −0.6 g Cd ha^{-1} yr^{-1}. In contrast, the balance of CPA$_{L6}$ and CPA$_{L12}$ were positive, with an accumulation of 1.65 g Cd ha^{-1} yr^{-1} and 1.92 g Cd ha^{-1} yr^{-1}, respectively (Table 6).	Excluding the scenario with leaching according to Eq. (8), only scenarios considering the enforcement of the EU regulation (L6 and L12) led to a negative balance, of the order of −0.2 to −0.6 g Cd ha^{-1} yr^{-1}. In contrast, the balance of CPA$_{L6}$ and CPA$_{L12}$ were positive, with an accumulation of 0.33 g Cd ha^{-1} yr^{-1} and 0.50 g Cd ha^{-1} yr^{-1}, respectively (Table 6).
		The Cd inputs can also be compared to those recently estimated for France by Belon et al. (2012) (Table 6).	The Cd inputs (1.89 g Cd ha^{-1} yr^{-1}) are very close to those recently estimated for France by Belon et al. (2012) (1.83 g Cd ha^{-1} yr^{-1}, Table 6).
		The mean fertilizer input which can be obtained from their work, is 1.02 g Cd ha^{-1} yr^{-1} at the national scale, while it was 2.84 g Cd ha^{-1} yr^{-1} in the CPA scenarios of this study.	The mean fertilizer input which can be obtained from their work, is 1.02 g Cd ha^{-1} yr^{-1} at the national scale, while it was 1.42 g Cd ha^{-1} yr^{-1} in the CPA scenarios of this study.
		When projecting the current practice into the future, the Cd balance that we calculated for France (+1.65 g Cd ha^{-1} yr^{-1} to +1.92 g Cd ha^{-1} yr^{-1}) is rather different to that simulated for the EU (27 + 1) by Six and Smolders (2014), which is clearly negative (−1.47 g Cd ha^{-1} yr^{-1}).	When projecting the current practice into the future, the Cd balance that we calculated for France (+0.33 g Cd ha^{-1} yr^{-1} to +0.50 g Cd ha^{-1} yr^{-1}) is rather different to that simulated for the EU (27 + 1) by Six and Smolders (2014), which is clearly negative (−1.47 g Cd ha^{-1} yr^{-1}).
		In some regions, the current Cd input by organic amendments could be reduced if the amount of nitrogen added were more precisely taken into account for the N fertilization calculation. According to simulations of scenarios including fertilization good practice, it is particularly the case in Brittany, were the excess of applied organic amendments (mainly slurry and manure) is 76% more than crop N requirements (Table S6). This also leads to an excess of N and P application to soils, which become available for leaching and coastal eutrophication (Ifremer, 2001). This is also the case in Auvergne and Limousin, where the current excess of organic amendment application is 25% and 20%, respectively.	In some regions, the current Cd input by organic amendments could be reduced if the amount of nitrogen added were more precisely taken into account for the N fertilization calculation. This would also decrease N excess available for leaching and coastal eutrophication (Ifremer, 2001). This is the case in Auvergne, Franche-Comté, Limousin and Pays de Loire, where there is a current excess of organic amendment from 3% to 25% (Table S6). However, in Bretagne, where the excess of applied organic amendments (mainly slurry and manure) is 76% more than crop N requirements, the reduction of Cd input from organic amendments would be compensated by the Cd inputs from P fertilizer applied on maize, which has relatively low N and high P requirements.
3. Results and discussion	3.3. Cadmium in crops	At the national level, the crops with the highest Cd inputs are sugar beet and rape (Table 7), which receive the highest P fertilization, while those which most export the metal, are sugar beet, forage maize and sunflower.	At the national level, the crops with the highest Cd inputs are sugar beet, grain maize and rape (Table 7), which receive the highest P fertilization, while those which most export the metal, are sugar beet, forage maize, sunflower and durum wheat.
		It should even slightly decrease in the GPEU$_{L6}$ scenario, and be quasi-stationary in the case of the EUR$_{L6}$ scenario.	It should decrease only in the scenarios considering the enforcement of the EU regulation limiting Cd in P fertilizers. However, this decrease would be slow in all the cases.
4.		If current cultivation practices are continued, the average Cd content in	If current cultivation practices are continued, the average Cd content in

Conclusions		French soils under annual crops will increase by about 15% by the end of the next century.	French soils under annual crops will increase by about 3 to 5% by the end of the next century.
		The cause of this increase is the input of Cd with P fertilizer applications, which represents around 85% of Cd inputs in soil, and which is nearly twice the Cd outputs by leaching and crop offtake.	The cause of this increase is the input of Cd with P fertilizer applications, which represents around 74% of Cd inputs in soil, and which corresponds to the Cd outputs by leaching and crop offtake.
		The main reasons of this contradiction are 1) a higher rate of P application in France than in Europe, 2) a higher Cd content in the French P fertilizers compared to the European ones and 3) lower Cd leaching in French soils, the leaching rate calculated in the study at the European level probably being overestimated.	The main reasons of this contradiction are a higher Cd content in the French P fertilizers compared to the European ones and a lower Cd leaching in French soils, the leaching rate calculated in the study at the European level probably being overestimated.
		In France, P applications on annual crops are excessive and could be reduced by about 50%, while still satisfying the crop requirements.	In France, P applications on annual crops are excessive and could be reduced by about 12% to 25%, while still satisfying the crop requirements.
		Assuming current excessive P fertilization, the enforcement of a regulation limiting Cd content in P fertilizers, as by proposed the European Union, would lead to a clearly lesser increase in soil Cd, of between 1.6% and 3.9% after 100 years. Only the combination of P fertilization good practices and of a regulation limiting Cd in P fertilizers would lead to a decrease of Cd in soil, of between 3.0% and 5.2%, after a century of conventional agriculture.	Assuming current excessive P fertilization, the enforcement of a regulation limiting Cd content in P fertilizers, as by proposed the European Union, would lead to a slight decrease in soil Cd, of between 1.6% and 3.8% after 100 years. The combination of P fertilization good practices and of a regulation limiting Cd in P fertilizers would lead to a decrease of Cd in soil, of between 3.0% and 5.2%, after a century of conventional agriculture.

References

AGRESTE. Agreste les Dossiers n° 21 - Enquête Pratiques culturales 2011. Ministère de l'Agriculture, de l'Agroalimentaire et de la Forêt, Paris, 2014.

DOI of original article: https://doi.org/10.1016/j.scitotenv.2018.05.225

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Science of the Total Environment 639 (2018) 1440–1452

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Cadmium mass balance in French soils under annual crops: Scenarios for the next century

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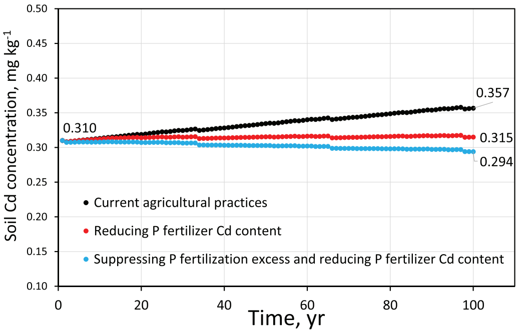
HIGHLIGHTS

- In France, P fertilizer applications represent around 85% of the soil Cd inputs.
- Maintaining current cultivation practices, soil Cd content would increase by *ca* 15%.
- Lessening both P fertilizer application and Cd content would reduce soil Cd content.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 6 March 2018
Received in revised form 17 May 2018
Accepted 18 May 2018
Available online 27 May 2018

Editor: Xinbin Feng

Keywords:
Agricultural practices
Phosphorus fertilizer
Cadmium leaching
European regulation

ABSTRACT

Human populations are threatened by chronic exposure to the Cd accumulated in foods after being taken up from soils by crops. To decide whether and to what extent it is necessary to reduce the Cd content in cultivated soils, one needs to understand and predict its evolution. We therefore simulated the Cd mass balance in the soils under annual crops in France and in its 22 regions for the next century, following six scenarios of agricultural practices or regulatory conditions. If current cultivation practices are maintained, the average Cd content would increase by about 15% after a century, due to the input of Cd with P fertilizer applications. This represents around 85% of the soil Cd inputs and is nearly twice the Cd output caused by leaching and crop offtake. These results conflict with those recently obtained at the European level, due to three factors: the higher rate of P application in France than in Europe, a higher Cd content in the P fertilizers applied in France and a lower Cd leaching in French soils. Strict application of the good practices for P fertilization would stabilize the future soil Cd content at its present level. Assuming the current excessive P fertilization, the enforcement of a regulation limiting Cd content in the P fertilizers, as proposed by the European Union, would lead to a lesser increase in soil Cd, by 1.6% to 3.9% after a century. The combination of P fertilization good practices and Cd content limitation in P fertilizers would lead to a decrease in soil Cd content of between 3.0% to 5.2%. Organic agriculture would lead to an evolution of soil Cd content similar to that of conventional agriculture applying good practices. The accuracy of the mass balances could be ameliorated by a better assessment of Cd leaching.

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https://doi.org/10.1016/j.scitotenv.2018.05.225
0048-9697/

1. Introduction

Cadmium (Cd) is a trace metal which is highly toxic to living organisms. It is strongly retained in the human body, particularly in the kidneys (Järup and Åkesson, 2009). This makes low-level chronic exposures a serious threat to human health, increasing the risk of kidney failure and cancer (EFSA, 2012). In the non-smoking population, food is the main source of human exposure to Cd. The US Agency for Toxic Substances and Disease Registry (ATSDR, 2012) and the European Food Safety Authority (EFSA, 2012) have set recommended provisional safe intake limits for Cd. If the EFSA limit is considered (2.5 μg Cd per kg of body weight per week), the populations of Europe (2.04 μg Cd $kgbw^{-1}$ w^{-1}), USA and China are on average just below the limit, whereas those of Japan and much of the remaining world would be above. Referring to the US limit (0.7 μg Cd $kgbw^{-1}$ w^{-1}), nearly all the populations in the world would be overexposed to Cd in food (Clemens et al., 2013). The most recent study by ANSES (2011) estimated that in France, the weekly intake is 1.12 μg Cd $kgbw^{-1}$ w^{-1} for adults and 1.68 μg Cd $kgbw^{-1}$ w^{-1} for children. The EFSA limit was exceeded for 0.6% of French adults and 14.9% of children. As a consequence, ANSES (2011), as well as the European Commission (Borg, 2014) recommend that efforts should continue to reduce dietary intakes of Cd.

In Europe, the EFSA (2012) estimated that grains and grain products (26.9%), vegetables and vegetable products (16.0%) and starchy roots and tubers (13.2%) had the greatest impact on dietary exposure to Cd. Similar origins were found for the French dietary intake of the metal (ANSES, 2011). Cadmium in vegetables comes from cultivated soils, through plant root absorption and translocation to the harvested organs. Most of the soil Cd originates from contamination due to human activities. The sources of contamination are phosphate fertilizers, atmospheric deposition, organic and mineral amendments. As atmospheric deposition from industrial emissions have decreased since the mid-1960s, Cd from P fertilizer is now the major input into cultivated soils (Six and Smolders, 2014).

That is why since 2003, European authorities have tried to set up a regulation limiting the Cd content in P fertilizers in order to prevent further accumulation in soils. The maximum limits would be established in three successive steps of 60, 40 and 20 mg Cd kg^{-1} P_2O_5 (DG Enterprise, 2003). According to the latest regulation proposal (Union Européenne, 2016), the dates at which these maximum limits would apply would be, respectively, the entry into force of the regulation (60 mg Cd kg^{-1} P_2O_5), 3 years (40 mg Cd kg^{-1} P_2O_5) and 12 years (20 mg Cd kg^{-1} P_2O_5) after the entry. However, such a regulation is still under discussion, as it would favour suppliers of low-Cd phosphate rocks whereas suppliers of high Cd phosphates would need to invest in decadmiation technology. The cost of raw materials for fertilizer manufacturers in the EU would increase, and the additional cost would be passed on to the farmers.

One element of this discussion is the study by Six and Smolders (2014). Carried out according to a method close to that already used in the Australian environment (de Vries and McLaughlin, 2013), it consisted in simulating Cd mass balance in European agricultural soils under current Cd fluxes, in order to predict the change in Cd content after 100 years of potato or wheat cultivation. The Cd mass balance was computed for the ploughed horizon, where the Cd inputs were from the atmospheric deposition, the P fertilizers, as well as from the mineral (liming) and organic (manure, sludge) amendments. The Cd outputs were those from leaching and crop offtake.

Taking an average scenario, Six and Smolders (2014) predicted that soil Cd concentrations in the EU would decrease by 15% over the next 100 years. The regional trends ranged between a 15% increase (e.g. Spain) and an 18% decrease (e.g. Germany). The authors related this variation to differences in soil pH, precipitation excess and fertilizer application rates. A sensitivity analysis of the balance model showed that soil pH was the most important factor of the long-term change in soil Cd, because of its impact on Cd leaching, followed by the initial soil Cd and organic C concentrations and by Cd content of the P fertilizer.

Cadmium mass balance in French agricultural soils was not simulated in the work of Six and Smolders (2014), in contrast to that of five other European countries (SE, DE, ES, UK, CZ). French agriculture has the first place in Europe in terms of utilised agricultural area (UAA) and standard production (16% of the EU-28, Eurostat (2013)). Moreover, it is based on specific and variable soils, crops and agricultural practices. Indeed, the mean soil pH in $CaCl_2$ is 6.5 and is quite different to the mean European value (5.8). The organic C content in French soils is 1.6%, which is 0.9% below the mean European content. Moreover, the Cd content of the P fertilizers, i.e. 51 mg Cd (kg P_2O_5)$^{-1}$ the second highest in European countries, is clearly above the European mean value (36 mg Cd (kg P_2O_5)$^{-1}$) and ten times greater than that of Swedish P fertilizers (Nziguheba and Smolders, 2008). The crops cultivated in France are also much more diverse than the sole potato and wheat crops considered by Six and Smolders (2014). The main crops (> 200,000 ha yr^{-1}) in France are wheat, rape, barley, grain maize, forage maize, sunflower, durum wheat, sugar beet and triticale. The mean P application rates on wheat and potato crops in France are 53 kg P_2O_5 and 84 kg P_2O_5 respectively, while those used at European level are 21 P_2O_5 and 45 P_2O_5, respectively (Six and Smolders, 2014).

In addition, the soil, climate and crops also strongly vary from one French region to another. For instance, pH ranges from 5.75 in the Limousin area to 7.63 in that of Provence-Alpes-Côte d'Azur, whilst soil Cd content varies from 0.19 mg Cd kg^{-1} in Aquitaine to 0.50 mg Cd kg^{-1} in Poitou-Charentes and organic C is 1.0% in Languedoc-Roussillon, but 2.6% in Brittany. While the precipitation excess is 357 mm on average in France (to be compared to the 200 mm taken by Six and Smolders (2014) for Europe), it varies from 179 mm in the Centre region to 750 mm in Franche-Comté. Although wheat is cultivated nearly everywhere, the crop rotations are quite different from one region to another. As an example, soils from Alsace are nearly all under grain maize monocropping, while eight different crops are commonly cultivated in the Midi-Pyrénées.

As a consequence, we simulated the Cd balance in French soils under annual crops at the level of the whole country and also at that of its 22 regions, for the coming 100 years. The regional simulations not only allowed an evaluation of the variability behind the simulations at the national level, but also provided information that could be used by the stakeholders to modify the agricultural techniques, which would take into account the local agro-pedo-climatic conditions. We also took the opportunity of this work to evaluate the contribution of amendments such as urban compost, industrial sludge and effluents on the Cd balance.

The Cd mass balance was simulated according to six scenarios affecting the P fertilizers' composition and application rates. Four scenarios were based on conventional agriculture practices, while two others supposed a complete conversion of French agriculture to organic farming. This article presents the trends in soil Cd content for the next century according to the scenarios, but also those of the different mass balance items.

2. Materials and methods

2.1. Simulation scenarios

The mass balance simulations were run according to six scenarios:

- CPA scenario: both the current P application rates of each crop as well as the Cd content in the fertilizer are maintained for the next 100 years.
- GPPA scenario: P applications are made according to good practices, i.e. referring to the method recommended by the French Committee

for the Study and Development of Reasoned Fertilization (COMIFER).

- EUR scenario: P fertilizers are applied at current rates as in the CPA scenario, but an EU regulation limiting their Cd content is enforced at the start of the simulation, progressively reducing the fertilizer Cd content.
- GPEU scenario: EU regulation are enforced and P fertilizer is applied according to good practices.
- OA scenario: French agriculture is completely converted to organic agriculture.
- OAEU scenario: similar to OA, with the enforcement of an EU regulation limiting Cd concentration in P fertilizers.

Organic agriculture was simulated based on conventional agriculture, with a differentiation similar to that adopted by Muller et al. (2017). The crop yields were reduced by 25%. Only manure and slurry were applied as organic amendments, with 10% smaller amounts, corresponding to the reduction in livestock production. Fertilization was carried out according to good practice (as in GPPA and GPEU scenarios), with rock phosphate as P source. A legume crop, i.e. field pea, was introduced into the rotation defined for conventional agriculture every 5 years. This way of simulating organic farming practices is a simplification. Indeed, the crops in the rotations should probably be more modified, following the deep structural changes in production systems that conversion to organic farming implies. These structural changes are difficult to conceive now, especially since they should be adapted to each pedo-climatic context i.e. to each region. This is why OA and OAEU scenarios were applied only at the national scale and not at the regional one.

2.2. Description and parameterization of the mass-balance model

2.2.1. Mass balance model

The Cd content in the topsoil is calculated annually according to

$$[\mathrm{Cd}]_{Soil,n} = [\mathrm{Cd}]_{Soil,n-1} + \frac{(Q_{in} - Q_{out}) * 1000}{M_{soil}} \quad (1)$$

so the soil Cd concentration over the year *n* ($[\mathrm{Cd}]_{Soil,\ n}$, mg kg^{-1}) is the result of that in year *n*-1 ($[\mathrm{Cd}]_{Soil,\ n-1}$) and of the balance between the input (Q_{in}, g ha^{-1}) and output (Q_{out}) of Cd in the considered layer, whose mass is M_{soil} (kg ha^{-1}).

Q_{in} is obtained by summing the quantity of Cd (g ha^{-1}) brought by atmospheric deposition (Q_{atm}), P fertilizer applications (Q_{pho}), liming (Q_{lim}) and organic amendments (Q_{org}):

$$Q_{in} = Q_{atm} + Q_{pho} + Q_{lim} + Q_{org} \quad (2)$$

The Cd outputs consist in the sum of the quantity of Cd leached out of the soil layer (Q_{lea}) and the crop offtake (Q_{crop}):

$$Q_{out} = Q_{lea} + Q_{crop} \quad (3)$$

The Cd mass balance was calculated for the mass of soil corresponding to the upper 25 cm of the soil on a 1 ha surface area. This corresponds to the ploughed layer of soils under annual crops. As in Six and Smolders (2014), the impact of surface runoff and erosion was not taken into account, as at each point of the cultivated area, Cd arriving from upstream was considered to compensate for Cd lost downstream.

The mass balance was calculated for 100 successive years for France as a whole and again for each of its 22 administrative regions. Each of these spatial units was considered to be covered by a unique virtual field with homogeneous soil and climate properties, represented by mean parameter values obtained from the real various soils. Each year, the mass balance was calculated according to the crop rotation defined for the spatial unit, the inputs and outputs being as far as possible specific to the given region (in fact, only Q_{atm} and Q_{lim} could not be regionalised, see below).

The mass balances were simulated with codes written with R software, version 3.2.3. through RStudio 1.0.136 version, the outputs being processed using Microsoft Excel® 2013.

2.2.2. Soil Cd content and other parameters

Knowledge of the initial soil Cd content is a pre-requisite to the balance modelling. This arithmetically affects the simulated future concentrations, both as the contributions of each balance item are added to it, but also because leached amounts and crop offtake (which are the two output items) are functions of soil Cd concentration (see below). Soil Cd content for France and each of the regions were provided by the soil analysis database for trace metals (BDETM) of the Gis Sol (Duigou and Baize, 2010) (http://www.gissol.fr/donnees/donnees-de-la-bdetm-2873). For each of the spatial units, the mean content was calculated using the data collected between 1990 and 2010 (Table 1). This long period was chosen in order to have a sufficient number of analyses, considering that the Cd content variation would be relatively small over that period.

Other necessary soil parameters such as pH (in water), organic C, clay, total carbonate and available (Olsen) P contents (Table 1) were obtained from the database of soil analyses (BDAT) of the Gis Sol (Saby et al., 2014) (http://estrada.orleans.inra.fr/geosol/), using the data from the most recent collection period, i.e. 2005–2009. For the prediction of Cd in soil solutions (see below), pH in water extracts (pH_{water}) was transformed into pH in 0.01 M $CaCl_2$ (pH_{CaCl2}), according to the relationship used by Six and Smolders (2014).

$$pH_{CaCl2} = pH_{water} - 0,54\ \left(R^2 = 0.88, n = 86\right) \quad (4)$$

The apparent soil density was taken to be 1.4 t m^{-3} (Bruand et al., 2004; COMIFER, 2016). The UAA was provided by AGRESTE, the statistical department of the French Ministry of Agriculture (http://agreste.agriculture.gouv.fr/). The values from the latest enquiry (2010) were used (Table S1).

Apart from the Cd content, all other soil parameters were considered as being stationary during the century in which the Cd balance was simulated. In particular, pH and organic C content, (which affect Cd leaching in the model, see 2.2.7) were considered to be at equilibrium in the scenarios of conventional agriculture, based on current liming and organic matter inputs. Organic agriculture is often considered to increase soil organic carbon. However, this belief might be based on analyses in farming systems in which organic carbon is imported (Leifeld and Fuhrer, 2010). In the organic agriculture system simulated here, there is no carbon input from outside. It is therefore unlikely that soil organic C content would increase when yield is reduced by 25%, as carbon fixation by the crops could be reduced in a similar proportion. Nevertheless, we considered that through its practices, organic agriculture maintained the soil organic carbon and the pH constant.

2.2.3. Inputs from P fertilizers

These were calculated by multiplying the weight of P fertilizer applied each year by the fertilizer's Cd content. The amount of P fertilizer applied (kg P_2O_5 ha^{-1}) was estimated in two ways, according to the scenario considered for the balance simulation. Namely the current application rate and the recommended application rate. The mean current dose applied to each crop in France and in each region was obtained from AGRESTE. In particular, national mean values were taken from AGRESTE (2014), while regional values were provided on demand by AGRESTE. These data resulted from the most recent enquiry carried out by the service in 2011 (Table S2).

Table 1
Mean soil characteristics used in the Cd balance simulations. W_{lea}: precipitation excess.

Spatial unit	W_{lea}	Total Cd	Clay	Total carbonate	pH		Organic C	P_2O_5 Olsen
	mm yr^{-1}	mg kg^{-1}	g kg^{-1}	g kg^{-1}	Water	$CaCl_2$ 0.01 M	g kg^{-1}	mg kg^{-1}
Alsace	350	0.27	237.84	72.47	7.36	6.82	13.81	87.53
Aquitaine	447	0.19	176.75	96.12	7.04	6.50	13.74	82.45
Auvergne	370	0.38	186.07	34.10	6.62	6.08	19.33	63.90
Bourgogne	326	0.33	265.20	156.89	7.37	6.83	16.33	70.43
Bretagne	378	0.21	172.23	7.35	6.36	5.82	25.63	118.69
Centre	207	0.25	223.96	65.47	7.11	6.57	12.50	75.89
Champagne-Ardenne	333	0.45	331.29	396.04	8.00	7.46	17.35	85.87
Corse	380	0.31[a]	155.16	6.91	6.66	6.12	10.71	47.38
Franche-Comté	750	0.50	298.32	67.95	6.67	6.13	19.36	58.76
Ile de France	179	0.29	186.70	44.74	7.39	6.85	11.16	90.33
Languedoc-Roussillon	363	0.29	217.10	207.07	8.02	7.48	10.11	45.99
Limousin	496	0.27	180.40	136.93	6.29	5.75	20.76	61.42
Lorraine	450	0.27	340.82	113.06	7.39	6.85	19.79	69.51
Midi-Pyrénées	384	0.27	220.13	88.07	6.84	6.30	12.60	80.21
Nord-Pas de Calais	287	0.43	182.61	48.49	7.71	7.17	12.49	121.10
Basse-Normandie	331	0.21	187.44	84.75	6.55	6.01	20.28	77.11
Haute-Normandie	315	0.33	152.70	12.85	7.06	6.52	12.17	78.31
Pays de Loire	254	0.22	169.66	29.36	6.56	6.02	15.46	95.90
Picardie	235	0.38	191.67	70.25	7.88	7.34	11.89	93.79
Poitou-Charentes	266	0.50	254.80	202.37	7.29	6.75	17.54	72.18
Provence-Alpes-Côte d'Azur	370	0.27	238.18	264.46	8.17	7.63	11.38	53.60
Rhône-Alpes	655	0.30	170.77	67.11	6.76	6.22	15.71	78.43
France	357	0.31	216.64	103.12	7.07	6.53	15.96	80.27

[a] Missing value replaced by national mean.

The recommended application rate (D_P), was calculated based on the COMIFER method (http://www.comifer.asso.fr/index.php/fr/):

$$D_P = P_{har} Y C_r - P_{org} \tag{5}$$

where P_{har} is the P content of the harvested part (kg P_2O_5 t^{-1}), whose weight is Y (t ha^{-1}) and C_r is a coefficient ranging from 0 to 3.7 according to the crop's requirement level, the soil content of available P and the frequency of P fertilizer application. P_{org} is the amount of efficient P added by organic amendments (COMIFER, 1995; COMIFER, 2009).

The regional and national crop yields (Y) were provided by AGRESTE (means of yields from 2011 to 2015, Table S3) and P_{har} by COMIFER (2009). C_r was taken from COMIFER (2009), according to a grid resulting from the combination of three classes of soil P content with three classes of crop requirement, considering that the P fertilizer was applied annually. As the soil available P mean content was high, (on average <10% below the upper limit (T_{imp}) for the crop with the highest P requirement (Table 1 and Table S4)), C_r was taken to be 1.5 for the crops with high P requirements (sugar beet, rape, potato) and 1 for the crops with low and medium P requirements (*i.e.* all the other crops). This fertilization should ensure the preservation of the soil's available P content. The contribution of organic amendments to P supply (P_{org}) was calculated for each crop according to

$$P_{org} = \sum_{i=1}^{n} m_i P_{org_i} K_{eq_i} \tag{6}$$

where m_i is the mass of organic amendment i (kg ha^{-1}), P_{org_i} is the amendment's P content (kg P_2O_5 kg^{-1}) and K_{eq_i} (≤ 1) is an equivalence coefficient of the efficiency of P in organic amendment, in comparison to P in mineral fertilizers. P_{org_i} was taken from Houot et al. (2014), while K_{eq_i} was taken from ARVALIS (2016) (Table S5). The estimation of m_i is given below.

The current fertilizer Cd content used in conventional agriculture was taken as 51 mg Cd (kg P_2O_5)$^{-1}$. This value was obtained from the analysis of 18 fertilizer samples (Nziguheba and Smolders, 2008). In the scenarios considering organic agriculture, rock-phosphate was the P source (Muller et al., 2017) and contained 47 mg Cd (kg P_2O_5)$^{-1}$. The method used to calculate this value is given in Supplementary Material (Table S7), based on data from McLaughlin et al. (1996) and Bech et al. (2010). In the scenarios where the EU regulation was supposed to be enforced, Cd concentration in the P fertilizer was the current concentration during the first three years (51 or 47 mg Cd (kg P_2O_5)$^{-1}$), 40 mg Cd (kg P_2O_5)$^{-1}$ in the nine subsequent years and 20 mg Cd (kg P_2O_5)$^{-1}$ from year 13 to year 100 (Union Européenne, 2016).

2.2.4. Inputs from organic amendments

The annual Cd input due to organic amendments was calculated according to

$$Q_{org} = R_N \sum_{i=1}^{n} m_{org_i} Cd_{org_i} K_{Cd_i} \tag{7}$$

m_{org_i} being the mass of organic amendment, Cd_{org_i} its Cd content and K_{Cd_i} a coefficient ranging from 0 to 1. R_N is a coefficient ($0 \leq R_N \leq 1$) limiting the excess of N introduced through organic amendments. This only applies in scenarios where fertilization is calculated according to good practices (see below). In conventional agriculture-based scenarios, the organic amendments considered were manure, slurry, sewage sludge, urban composts, industrial organic wastes, sludge and effluents from the agri-food industry and sludge and effluents from other industries (non agri-food). In the scenarios based on organic agriculture, only manure and slurry were applied to respect the European Commisison (2008) specifications. In all the scenarios, this contribution was considered as stationary throughout the century (i.e. based on current amendment applications), as nothing in the literature indicates any clear variation in the application of these products in the future.

The amounts of manure and slurry produced yearly in each region were taken from FranceAgriMer (2015). They were divided by the UAA under annual crop and grassland of the corresponding region, in order to obtain the current amount applied each year per ha (Table S6). This calculation supposed that the amendments were applied locally, because of transportation costs. The amount of sewage sludge spread on agricultural soil in each French administrative department in 2015 was obtained from the Ministry of the Environment (http://assainissement.developpement-durable.gouv.fr/services.php), the regional and national values being derived by summing up the departmental quantities. The quantity of urban compost, made from domestic vegetal wastes and organic refuse, was calculated as follows.

The amount of domestic vegetal waste produced in each region was obtained from FranceAgriMer (2015). The mass of compost was deduced by considering that composting reduces the original waste mass by 50% (Lopez (2002); personal communication from Valterra composting plant) and that the part of domestic refuse in urban compost is 15.8% (Houot et al., 2014). The amounts of industrial organic waste, sludge and effluents from agri-food- and non agri-food industry produced in each region were found in Houot et al. (2014).

In scenarios where P fertilization was calculated according to good practice, the amount of N required by the crop was also calculated basing on the COMIFER method (COMIFER (2011); see also Supplementary Material). Consequently, when the N added by organic amendment exceeded the crop requirement, the quantity of amendments applied was reduced to balance the N supply with the crop requirement and to avoid N leaching. This was operated through R_N calculated as the ratio of N crop requirement (kg N ha^{-1}) to N provided by the organic amendments (if $R_N \geq 1$, $R_N = 1$).

The Cd and N contents (Table S5) in organic amendments were obtained from Houot et al. (2014). K_{Cd_i} was 1 for all the organic amendments, except for manure. In this case, K_{Cd_i} was 0.18, in order to take into account the fact that Cd input from straw in manure, was in fact a restitution of Cd removed with the straw used locally for animal litter (Supplementary Material).

2.2.5. Inputs from liming

To estimate the amount of Cd brought through the application of calcium magnesium amendments, we used the delivery of basic amendments to metropolitan France from 2011 to 2015, as provided by the ANPEA (http://www.anpea.com/livraisons-de-fertilisants/livraisons-d-amendements.html). Summing carbonate and lime, the annual delivery was on average 945,000 t CaO (Table S8). The Cd content of the amendment was taken to be 0.35 mg Cd (kg CaO)$^{-1}$ (Six and Smolders, 2014). This value is consistent with the fact that calcareous rocks generally contain relatively high amounts of Cd (Baize et al., 1999; Quezada-Hinojosa et al., 2009). Considering the 17,556,765 ha under annual crop in France, the average liming is 54 kg CaO ha^{-1} yr^{-1}, which brings 0.02 g Cd ha^{-1} yr^{-1}. This value will vary from one region to another, depending on the soil properties, however regional values were not available.

2.2.6. Atmospheric deposition

According to Ilyin et al. (2016), the atmospheric deposition of Cd in France was 18 g Cd km^{-2} on average in 2014. The value of 0.2 g Cd ha^{-1} was used here for France as a whole, as well as for each of its 22 regions. This is within the range of the values recorded by Six and Smolders (2014) (0.1 to 0.3 g Cd ha^{-1}).

2.2.7. Output by leaching

This item was estimated according to the method used by both de Vries et al. (2011) and Six and Smolders (2014), i.e. multiplying the amount of water leaving the topsoil (W_{lea}, mm yr^{-1}) by the Cd concentration in the soil solution ($[Cd]_w$, mg Cd L^{-1}):

$$Q_{lea} = 10\ W_{lea}\ [Cd]_w \tag{8}$$

W_{lea} being considered as the annual precipitation excess. This was provided by Météo-France as normal values for the period 1981–2010. The latter were computed using the SAFRAN-ISBA-MODCOU hydrometeorological model (Habets et al., 2008) at each node of a 8 km square grid. The nodal values were aggregated to compute the regional and national mean values (Table 1).

To estimate $[Cd]_w$, we used the model adopted by Six and Smolders (2014) for their prediction of Cd balance at the European level:

$$[Cd]_w = \frac{[Cd]_s}{K_D} \tag{9}$$

with

$$\log(K_D) = -0.94 + 0.51pH_{CaCl2} + 0.79\ \log(C_{org})\ \left(n = 151, \mathrm{R}^2 = 0.71\right) \tag{10}$$

C_{org} being the soil organic C content (% mass) and $[Cd]_s$ being the Cd on the soil solid phase, taken as the total soil Cd content.

Cadmium concentration in percolating water has rarely been measured in the field under cultivated soils. Degryse and Smolders (2006) compared Cd concentrations in percolating and in pore water at the same depth (0.7 m) in three profiles of cultivated soils (two slightly contaminated and one control). The ratios of Cd concentration in pore water to those in the corresponding leachates were on average 1.4, 0.9 and 1.7 after 18 months' of sampling. The highest value was that of the control soil containing 0.18 mg Cd kg^{-1} in the upper horizon. Bengtsson et al. (2006) measured Cd concentrations and fluxes in percolating soil water and surface run-off on arable land, in an experimental farm from Northern Sweden. They collected soil water for five years, from four sampling sites differing in soil type and agricultural practice. There were three to four replicated water sampling devices (suction cup lysimeters) collecting water at three depths (20, 50 and 80 cm). Topsoils contained 0.10–0.11 mg Cd kg^{-1}. The Cd concentration measured in the solutions at the different depths was on average 12 times lower than that estimated with Eq. (9) (Table S9). The authors estimated the average Cd flow through percolation and run-off to be 0.35 g Cd ha^{-1} yr^{-1}, which is much lower than the 2.56 g Cd ha^{-1} yr^{-1} average leaching calculated at the European level by Six and Smolders (2014) using Eq. (9). More recently, Cambier et al. (2014) and Filipović et al. (2016) have published data from the QualiAgro field experiment in Feucherolles, near Paris (France), which belongs to the French national SOERE-PRO network. They measured Cd water concentrations and fluxes from nearly six years of lysimeter sampling at a 45 cm depth, from three plots cultivated with maize and wheat. One was a control plot, the other two received either sewage sludge compost or municipal waste compost. The Cd concentration measured in the percolating water (Cambier et al., 2014) was 15 times lower than that predicted by Eq. (9). The Cd leached ranged between 0.11 and 0.14 g Cd ha^{-1} yr^{-1} (Filipović et al., 2016), i.e. about 12 times lower than that estimated with Eq. (8) (Table S10). We also obtained the data from another field experiment of the SOERE-PRO network, namely the PRO'spective experiment located in Alsace, near Colmar. Cadmium concentrations and fluxes were measured at 45 cm for 5.5 years using two lysimeters per plot, in six plots cultivated with grain maize, winter wheat, sugar beet and spring barley. One of the plots was a control plot without amendment, the five others receiving either sewage sludge, composted sewage sludge, composted biowaste, bovine manure and composted manure. On average, the measured Cd concentrations in soil solutions were 3.5 times lower than those simulated, while the measured Cd fluxes were 5.1 times lower (Table S11).

As there is a strong probability that the leached Cd (Q_{lea}) would be overestimated using Eq. (8), the latter was modified so the Cd leaching could roughly adjusted to the results of the field trials of Cambier et al. (2014) and Filipović et al. (2016):

$$Q_{lea} = 10\ W_{lea}\ [Cd]_w/12 \tag{11}$$

Because the results of the above-mentioned field trials could represent a situation where leaching is extremely reduced, another version of the model was implemented with a leaching rate falling empirically between the two previous ones, which is closer to that of Bengtsson et al. (2006) and to the PRO'spective experiment, i.e. six times lower than that predicted by the formalism of Six and Smolders (2014):

$$Q_{lea} = 10\ W_{lea}\ [Cd]_w/6 \tag{12}$$

Three balance models, differing in the Cd leaching equation were therefore run for each of the scenarios (see 2.1). When simulated with 12 and 6 times reduced leaching (Eqs. (11) and (12)), the scenarios are noted respectively with L12 and L6 as a subscript.

2.2.8. Output by crop offtake

Cadmium taken off by the harvested crop (Q_{crop}) was estimated by (Six and Smolders, 2014)

$$Q_{crop} = TF[Cd]_s Y \tag{13}$$

where the transfer factor, *TF* is given by

$$TF = \frac{[Cd]_{crop}}{[Cd]_s} \tag{14}$$

TF values were obtained from the literature for most of the crops (Table 2), as far as possible for French conditions. Details on the way they were calculated are given in Supplementary Material.

The crops were changed annually according to a rotation defined for each spatial unit (region or France as whole), as follows. The crops representing at least 3 to 5% of the UAA under annual crops (AGRESTE, 2014) were included in the rotation in conventional agriculture. The crop with the smallest UAA was considered to be the least cultivated one and to appear only once in the rotation. The number of times each of the other crops would appear in the rotation was deduced from the ratio of their UAA to that of the least-cultivated crop. The crops of the rotations and the number of times they appeared in the rotations are given in Table 3, while the rotations are given in Supplementary Material.

2.3. Sensitivity analysis

A sensitivity analysis was carried out using R software in order to evaluate which of the 14 parameters varying between the spatial units had the greatest impact on the predicted variation of soil Cd content after 100 years (V_{Cd}, %).

$$V_{Cd} = \frac{[Cd]_{s,\ t=100} - [Cd]_{s,t=0}}{[Cd]_{s,t=0}} 100 \tag{15}$$

in which $[Cd]_{s,\ t=0}$ and $[Cd]_{s,\ t=100}$ are the soil Cd contents at the start and at the end of the Cd balance simulation, respectively.

The analysis was carried out on scenario GPPA for wheat monocropping. The parameters were sampled using random sampling of uniform distributions of the parameters (Saltelli et al., 2004), with extreme values corresponding to those of the spatial units. The sample size was 10,000 (Six and Smolders, 2014). Factors were ranked according to ANOVA F values and standardized regression coefficients.

Table 2
Cadmium transfer factor (TF) used to estimate the Cd offtake of the different crops. The method used for their calculation is given in Supplementary Material.

Crop	TF
Barley	0.24
Durum wheat	0.47
Field pea	0.13
Forage maize	0.69
Grain maize	0.11
Potato	0.083
Rape	0.14
Sugar beet	0.41
Sunflower	1.25
Triticale	0.15
Wheat	0.15

3. Results and discussion

3.1. Future trends in French soil Cd content

When France was taken as a whole, the future evolution of Cd content in arable soils was simulated both by using the mean parameters representing the whole country and, on the other hand, by averaging the evolutions of Cd contents in the soil of each of the 22 regions. Both methods converged towards similar results (Table 4, Table S13).

In the case of simulations with the highest leaching rate (Eq. (8)), if current P fertilization practices are maintained in the future, i.e. when considering the CPA scenarios, the soil Cd content should slightly decrease ($V_{Cd} = -6.3\%$) after 100 years (Table 4). Alternatively, if suddenly, farmers strictly adopted the best practice for P application, as simulated in the GPPA scenario, the P fertilizer rates would be half those of current application rates, leading to a reduction of 19.7% of the Cd content in soils after 100 years, again supposing Cd leaching at its highest level. Enacting the EU regulation limiting Cd content in P fertilizers (EUR scenario) would cause a decrease of 18.4% in the soil Cd content. Considering the GPEU scenario, i.e. the combination of application of fertilization good practice and the enforcement of EU regulations, the simulations predicted $V_{Cd} = -24.4\%$. Scenarios OA and OAEU, supposing organic agriculture practices, predicted decreases in Cd content of 20.0% and 23.4% respectively. All scenarios with the highest leaching rate therefore predicted a reduction in Cd content in soils after a century. This result is consistent with that found by Six and Smolders (2014) at the European level.

However, simulations with 12 times lower leaching (Eq. (11)) gave very different predictions. When considering the CPA_{L12} scenario, the soil Cd content should increase by 17.4% after 100 years (Table 4). Scenarios $GPPA_{L12}$ and EUR_{L12} predicted small increases in soil Cd content, of 2.2% and 3.9%, respectively. Considering scenario $GPEU_{L12}$, the Cd concentration in French soils would be reduced by 3.0% after a century, while in the hypothesis of organic agriculture, scenarios $OA_{\ L12}$ predicted a slight increase of 2.3% and in the case of scenario $OAEU_{\ L12}$ a small decrease (−1.5%).

In the case of an intermediate Cd leaching rate (Eq. (12)), CPA_{L6} predicted an increase of 15.0% in soil Cd content. According to $GPPA_{L6}$, soil Cd content would not vary ($V_{Cd} = -0.1\%$) after 100 years and slightly increase according to EUR_{L6} ($V_{Cd} = 1.6\%$). Scenario $GPEU_{L6}$ predicted a decrease of 5.2%, while in the context of organic agriculture, scenarios $OA_{\ L6}$ predicted no variation and $OAEU_{\ L6}$ a small decrease (−3.7%).

It must be noted that whatever the scenario, the Cd content variation would hardly be detectable in the first 20 years, as it would be <5% (Table 4), a variation which is generally well below the uncertainty of a conventional soil analysis at the plot scale (Wopereis et al., 1988).

The highest Cd leaching rate according to Eq. (8) is unlikely, as its modelling supposes that the replenishing of the percolating water with Cd by the solid phase is instantaneous in all drainage episodes. However, Cd leaching is limited by the desorption process from the soil solid phase (Blume et al., 2016; Selim et al., 2013). This is supported by the results of Bengtsson et al. (2006); Cambier et al. (2014); Degryse and Smolders (2006), Filipović et al. (2016) and of the PRO'spective experiment mentioned above to justify Eq. (11). As the soils studied in the works of Cambier et al. (2014) and Filipović et al. (2016) had properties (pH, organic C, total Cd, Table S10) close to those of the French mean soil used in our balance scenarios at the national level (Table 1), it is reasonable to consider that the lixiviation according to Eq. (11) is more realistic than that estimated with Eq. (8). These considerations also suggest that the lixiviation calculated in the simulation of the soil Cd balance at the European level by Six and Smolders (2014) could be overestimated.

Bases on the fact that dividing the maximum leaching rate (Eq. (8)) by 6 or 12 give close predictions, CPA_{L6} would be more reliable than CPA in predicting the trend of soil Cd content in a future where “business as

Table 3
Number of occurrences of each crop in the rotations, for each region and for France, for conventional agriculture. No data means no occurrence. For organic agriculture, eight field pea crops were added to the rotation simulated at national level.

Spatial unit	Barley	Durum wheat	Forage maize	Grain maize	Potato	Rape	Sugar beet	Sunflower	Triticale	Wheat
Alsace			1	11			1			4
Aquitaine			1	5				1		1
Auvergne	1		2	2		1		1	2	6
Bourgogne	6		1	1		6		1	1	10
Bretagne	1		6	2		1			1	6
Centre	3	1		2		4		1		8
Champagne-Ardenne	6		1	1		4	2			9
Corse	3		2	9					2	1
Franche-Comté	4		2	4		4		1	1	9
Ile de France	2			1		2	1			6
Languedoc-Roussillon	2	12				1		5	1	1
Limousin	2		5	1					5	4
Lorraine	2		1			2				3
Midi-Pyrénées	2	2	1	3		1			1	5
Nord-Pas de Calais	2		3		2	1	2			10
Basse-Normandie	1		5			1				6
Haute-Normandie	2		3		1	4	1			13
Pays de Loire	1		7	3		1		1	1	9
Picardie	2		1	1	1	3	3			12
Poitou-Charentes	2	1	1	4		2		4	1	9
Provence-Alpes-Côte d'Azur	3	13		1		1		2	1	2
Rhône-Alpes	2		3	6		1		1	1	6
France	4	1	3	4		4	1	2	1	12

usual" would be the way to manage annual crops. In this case, topsoil Cd content would increase by around 15% after a century.

Scenario $GPPA_{L6}$ predicts no variation in soil Cd content over the next century. However, this scenario gives more an assessment of the weight of fertilization practices on the soil Cd content (by comparison with the CPA scenarios) than a realistic prediction. Indeed, it seems very unlikely that all the farmers would simultaneously and immediately reduce their P applications by around 50%, strictly applying the COMIFER (1995) recommendations. These recommendations have been promoted for two decades with progressive but incomplete success, as shown by the excess of P fertilization that was suggested both by our calculations (Table 5) and those of UNIFA (2014). Moreover, if the application of mineral P fertilizer did indeed declined between 1989 and 2008, its usage has been steady since then (UNIFA, 2014).

The EUR scenarios, which predict a slight increase in soil Cd content, appear more realistic, as the project of a regulation reducing Cd in P fertilizers has existed at least since 2003 (DG Enterprise, 2003) and especially as the European Commission produced a regulation proposal in 2016 (European Commisison, 2016). What makes this scenario uncertain is the fact that the decadmiation processes are not yet operational, although several Cd removal methods are available. These methods are the calcination of phosphate rock or the decadmiation of phosphoric acid by co-crystallisation, sulphide precipitation, ion exchange and solvent extraction (Fertilizers Europe, 2014). Decadmiation of phosphate rock could be done by calcination at 850–1000 °C. However, this is energetically expensive and produces less reactive phosphate rock (Fertilizers Europe, 2014), whose P content might be less available for crop nutrition. These technologies have not reached the industrial application level, but their additional cost have been estimated from about \$10 (t P_2O_5)$^{-1}$ to more than \$100 (t P_2O_5)$^{-1}$ (Fertilizers Europe, 2014). Supposing the maximum increase in the P fertilizer price due to decadmiation of €100 (t P_2O_5)$^{-1}$, this would increase the cost of the P fertilization for wheat (53 kg P_2O_5 ha^{-1}) by €5.3. This cost increase represents approximately 0.3% of the wheat production cost (1640 € ha^{-1} in 2014, Carpentier (2014)). An increase in the fertilizer prices would encourage farmers to reduce their use by applying P fertilizers at rates closer to the crop P requirements, as recommended by COMIFER. However, the increase in the cost of the P fertilizer

Table 4
Evolution of Cd content in the ploughed layer (25 cm) of the French average soil under annual crops, according to different scenarios.

Scenario	Soil Cd content, mg kg^{-1}				Variation, %		
	Initial	10 yrs	20 yrs	100 yrs	10 yrs	20 yrs	100 yrs
CPA: Current P application rates	0.31	0.306	0.305	0.291	−1.1	−1.6	−6.3
CPA_{L6}: Same as CPA, with leaching rate/6	0.31	0.313	0.319	0.357	1.0	2.8	15.0
CPA_{L12}: Same as CPA, with leaching rate/12	0.31	0.314	0.320	0.364	1.2	3.3	17.4
GPPA: P application according to good practices	0.31	0.302	0.295	0.249	−2.7	−4.8	−19.7
$GPPA_{L6}$: Same as GPPA, with leaching rate/6	0.31	0.308[a]	0.309	0.310	−0.6	−0.5	−0.1
$GPPA_{L12}$: Same as GPPA, with leaching rate/12	0.31	0.309[a]	0.310	0.317	−0.4	0.0	2.2
EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.31	0.305	0.300	0.253	−1.5	−3.1	−18.4
EUR_{L6}: Same as EUR, with leaching rate/6	0.31	0.312	0.314	0.315	0.7	1.3	1.6
EUR_{L12}: Same as EUR, with leaching rate/12	0.31	0.313	0.315	0.322	0.9	1.7	3.9
GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.31	0.301	0.294	0.234	−2.8	−5.3	−24.4
$GPEU_{L6}$: Same as GPEU, with leaching rate/6	0.31	0.308	0.307	0.294	−0.7	−0.9	−5.2
$GPEU_{L12}$: Same as GPEU, with leaching rate/12	0.31	0.308	0.308	0.301	−0.5	−0.5	−3.0
OA: Organic agriculture	0.31	0.302	0.296	0.248	−2.6	−4.7	−20.0
OA_{L6}: Same as OA, with leaching rate/6	0.31	0.308 [a]	0.309	0.310	−0.5	−0.3	0.0
OA_{L12}: Same as OA, with leaching rate/12	0.31	0.309 [a]	0.311	0.317	−0.3	0.2	2.3
OAEU: Same as OA, with EU regulation limiting Cd in fertilizers	0.31	0.302	0.294	0.237	−2.7	−5.0	−23.4
$OAEU_{L6}$: Same as OAEU, with leaching rate/6	0.31	0.308	0.308	0.298	−0.6	−0.6	−3.7
$OAEU_{L12}$: Same as OAEU, with leaching rate/12	0.31	0.309	0.309	0.305	−0.4	−0.2	−1.5

[a] This minimum value is the consequence of the high crop offtake of sunflower in year 7, made visible by a scenario with little variation in Cd balance over the century.

Table 5
Amounts of P_2O_5 applied to annual crops in France and in its region according to current practices (CPA) scenarios and to good practice (GPPA) scenarios.

Spatial unit	CPA	GPPA	Variation
	kg P_2O_5 ha^{-1} $(100\,yrs)^{-1}$		%
Alsace	6566	3830	−42
Aquitaine	5472	2195	−60
Auvergne	4910	1439	−71
Bourgogne	5753	2245	−61
Bretagne	3764	2395	−36
Centre	5711	3994	−30
Champagne-Ardenne	7931	3851	−51
Corse	5371	4464	−17
Franche-Comté	6160	1788	−71
Ile de France	4827	3173	−34
Languedoc-Roussillon	5026	2828	−44
Limousin	3982	1075	−73
Lorraine	6038	1885	−69
Midi-Pyrénées	5295	1946	−63
Nord-Pas de Calais	5480	1763	−68
Basse-Normandie	4296	1180	−73
Haute-Normandie	5357	2797	−48
Pays de Loire	4281	690	−84
Picardie	5827	4097	−30
Poitou-Charentes	5155	2376	−54
Provence-Alpes- Côte d'Azur	4305	2728	−37
Rhône-Alpes	4730	2067	−56
France	5297	2458	−54

appears too small to make the GPEU scenario more plausible than the GPPA one.

Scenarios OA and OAEU are also quite unrealistic, as the total and rapid conversion of French agriculture to organic agriculture practices is very unlikely. However, it is probable that organic farming will progressively increase its share in the UAA (currently 5.29%) to join the European mean (6.69%) or even that of leading European countries (21.25% of the UAA in Austria, Eurostat (2017)). Organic agriculture would have similar consequences for soil Cd content to the application of fertilization good practices in conventional agriculture (scenarios GPPA and GPEU): only the reduction of Cd in P fertilizer, i.e. phosphate rock, would lead to a reduction in soil Cd content. However, because of the low solubility of P in phosphate rock, particularly in neutral to alkaline soils (Khasawneh and Doll, 1979), it cannot be excluded that the P fertilizer application in organic agriculture would be greater than in conventional agriculture, as determined by the crop's P requirements. It is therefore possible that the Cd balance in organic farming would be higher than in conventional agriculture.

In the different scenarios, the current crop rotations were generally repeated for a century, without taking into account that these could vary, due to changes in climate, social demand or economics in the future. Indeed, these changes are nearly impossible to predict. However, the OA and OAEU scenarios help in predicting the impact of crop rotation variations as they simulate an extreme modification of the farming practices, with a change in the rotations (introduction of a legume crop every 5 years) and consistent reduction of yield, and of fertilizer and amendment application. They show that the Cd balance does not consistently differ from that of conventional agriculture, because inputs and outputs simultaneously decrease in organic farming. The results of the OA and OAEU scenarios, compared to that of the other scenarios, indicate that changes in the future rotations should have less effect on Cd balance than any variation in the fertilizer Cd content would have.

Recently (European Parliament, 2017), the European Parliament amended the proposal concerning the rules on the making CE approved fertilising products available on the market (European Commisison, 2016). The time schedule fixing the time the Cd content in P fertilizers should be limited to 40 and 20 mg Cd $(kg\ P_2O_5)^{-1}$ has been extended from three and 12 years (after enforcement of the regulation) to six and 16 years, respectively. We compared the evolution of the soil Cd content in the scenarios involving the European regulation, in the case of moderate Cd leaching (EUR_{L6}, $GPEU_{L6}$ and $OAEU_{L6}$), for both the regulation variants (Table S14). Increasing the time lapses of application of the threshold contents resulted in a higher increase (EUR_{L6}: +0.5%) or a lower decrease ($GPEU_{L6}$: −0.2%; $OAEU_{L6}$: −0.1%) of soil Cd. However, the differences in V_{Cd} between the two regulation variants were small.

Variation in soil Cd content depends on the region considered (Table S15). Results from the Centre and Nord-Pas de Calais administrative regions can be used to illustrate the differences between regions (Fig. 1). In the Centre, scenario CPA_{L12} predicted an increase in the soil Cd content of 30.5% after a century, while in the Nord-Pas de Calais, it predicted only a 3.4% increase. Scenarios $GPEU_{L12}$ would conclude by a clear decrease in V_{Cd} in the Nord-Pas de Calais (−11.0%), which would not be the case in the Centre, where $GPEU_{L12}$ would lead to an 8.3% increase in Cd content.

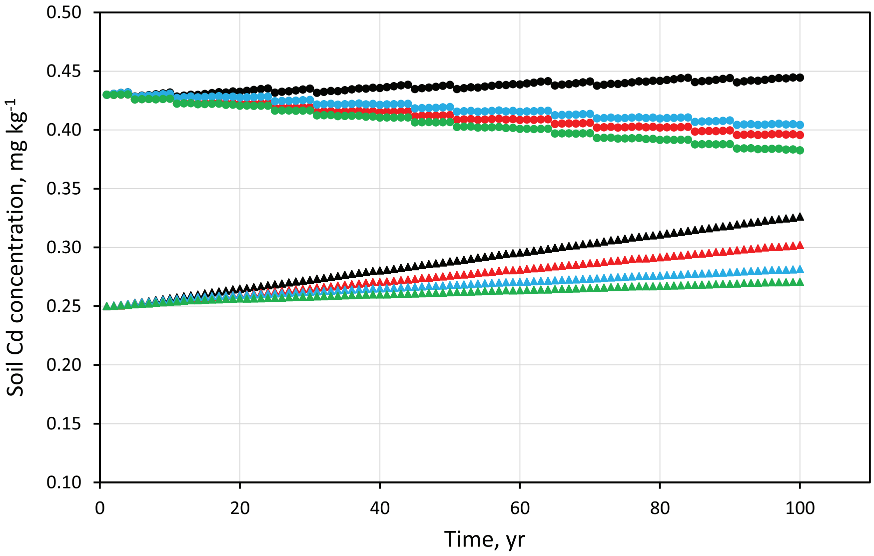

Fig. 1. Evolution of Cd concentration in the ploughed layer (25 cm) of soils under annual crops in the Centre (triangles) and Nord-Pas de Calais (circles) regions according to four scenarios with the lowest Cd leaching (Eq. (11)). Scenarios are CPA_{L12} (black), $GPPA_{L12}$ (red), EUR_{L12} (blue) and $GPEU_{L12}$ (green). Discontinuities in the Nord-Pas de Calais curves are due to the sugar beet offtake, resulting from a high yield (90.5 t ha^{-1}) and a relatively high TF (0.41). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

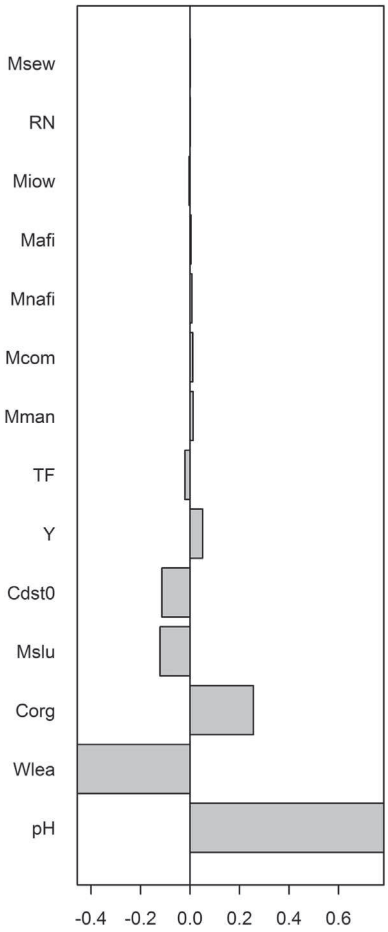

Fig. 2. Ranking of Cd balance variables based on their standardized coefficients in the multiple regression for variation in soil Cd content after one century (V_{Cd}). pH: Soil pH; Wlea: precipitation excess; Corg: Soil organic carbon; Mslu: mass of slurry; Cdst0: initial soil Cd content; Y: crop yield; TF: transfer factor; Mman: mass of manure; Mafi: mass of sludge and effluents from agri-food industry; Mcom: mass of urban compost; Mnafi: mass of sludge and effluents from other industries; Miow: mass of industrial organic waste; Msew: mass of sewage sludge; RN: crop nitrogen requirement.

The sensitivity analysis showed that the parameters which best explained the differences in V_{Cd} between regions were predominantly soil pH and precipitation excess and secondarily, soil organic C content, mass of applied slurry, initial soil Cd content, and crop yield (Fig. 2, Fig. S1). The first three parameters affect Cd leaching (Eqs. (8)–(10)), while initial soil Cd content affects Cd leaching (Eq. (9)), crop offtake (Eqs. (13) and (14)) and V_{Cd} itself (Eq. (15)). Crop yield simultaneously affects crop offtake (Eq. (13)), but also P fertilizer application rates (Eq. (5)). Among the organic amendments, slurry has the strongest impact on V_{Cd}, because it is the amendment whose application rate varies the most between regions.

3.2. Cadmium fluxes

Mean annual Cd fluxes in soils at the whole of France level are given in Table 6. Their annual variation over a century of cultivation is illustrated in Fig. 3. In the scenarios with current P application rates (CPA, CPA$_{L6}$, CPA$_{L12}$), the total inputs were 3.31 g Cd ha^{-1} yr^{-1}, P fertilizer application accounting for 2.84 g Cd ha^{-1} yr^{-1}, that is 85.5% of the Cd input in soil. Cadmium from atmospheric deposition accounted for 6.1% of the inputs, while the amendments each represented <2.0% of the Cd inputs (Table S16). This result indicates that the uncertainty of the estimation of the inputs by the various amendments has a minor impact on the Cd balance.

For the CPA scenario, the total outputs were 3.99 g Cd ha^{-1} yr^{-1} with Cd leaching of 3.00 g Cd ha^{-1} yr^{-1} (83.4% of the outputs) and 0.99 g Cd ha^{-1} yr^{-1} as crop offtake. In CPA$_{L6}$ and CPA$_{L12}$, leaching was 0.56 g Cd ha^{-1} yr^{-1} (53.8% of the outputs) and 0.28 g Cd ha^{-1} yr^{-1} (38.9% of the outputs), respectively. The leaching flux was between 2.7 and 3.0 g Cd ha^{-1} yr^{-1} when calculated according to Eq. (8). It was around 0.5 g Cd ha^{-1} yr^{-1} when calculated according to Eq. (12) and around 0.25 g Cd ha^{-1} yr^{-1} when assessed with Eq. (11). These last two leaching fluxes are much more consistent with the values measured in the field (Bengtsson et al., 2006; Filipović et al., 2016) than the first one. For a given leaching model, the amount of leached Cd varied little from one scenario to another, as it depends on parameters (precipitation excess, pH, organic C, Cd content) which do not or very slowly change during the balance simulation time. For similar reasons, in scenarios based on conventional agriculture, crop offtake varied little, at around 1 g Cd ha^{-1} yr^{-1}, whatever the leaching rate. However, when crop yields decreased, as in organic agriculture (−25% compared to

Table 6
Estimated mean cadmium fluxes (g Cd ha^{-1} yr^{-1}) in the ploughed layer (25 cm) of the mean French soil under annual crops for different scenarios.

Scenario	Inputs					Outputs			Balance
	P fertilizers	Organic amendments	Liming	Atmosphere	Total	Leaching	Crop offtake	Total	
CPA: Current P application rates	2.84	0.25	0.02	0.20	3.31	3.00	0.99	3.99	−0.68
CPA$_{L6}$: Idem CPA, with leaching rate/6	2.84	0.25	0.02	0.20	3.31	0.56	1.10	1.66	1.65
CPA$_{L12}$: Idem CPA, with leaching rate/12	2.84	0.25	0.02	0.20	3.31	0.28	1.11	1.39	1.92
GPPA: P application according to good practice	1.07	0.25	0.02	0.20	1.54	2.78	0.91	3.69	−2.15
GPPA$_{L6}$: Idem CPA, with leaching rate/6	1.07	0.25	0.02	0.20	1.54	0.52	1.02	1.54	0.00
GPPA$_{L12}$: Idem CPA, with leaching rate/12	1.07	0.25	0.02	0.20	1.54	0.26	1.03	1.29	0.25
EUR: Current P application rates with EU regulation limiting Cd in fertilizers	1.27	0.25	0.02	0.20	1.74	2.82	0.93	3.75	−2.01
EUR$_{L6}$: Idem CPA, with leaching rate/6	1.27	0.25	0.02	0.20	1.74	0.53	1.04	1.56	0.18
EUR$_{L12}$: Idem CPA, with leaching rate/12	1.27	0.25	0.02	0.20	1.74	0.27	1.05	1.31	0.43
GPEU: P application according to good practice with EU regulation limiting Cd in fertilizers	0.47	0.25	0.02	0.20	0.94	2.71	0.89	3.60	−2.66
GPEU$_{L6}$: Idem GPEU, with leaching rate/6	0.47	0.25	0.02	0.20	0.94	0.51	1.00	1.50	−0.56
GPEU$_{L12}$: Idem GPEU, with leaching rate/12	0.47	0.25	0.02	0.20	0.94	0.26	1.01	1.26	−0.32
OA: Organic agriculture	0.83	0.08	0.02	0.20	1.13	2.77	0.55	3.32	−2.19
OA$_{L6}$: Same as OA, with leaching rate/6	0.83	0.08	0.02	0.20	1.13	0.52	0.60	1.12	0.01
OA$_{L12}$: Same as OA, with leaching rate/12	0.83	0.08	0.02	0.20	1.13	0.26	0.61	0.87	0.26
OAEU: Same as OA, with EU regulation limiting Cd in fertilizers	0.40	0.08	0.02	0.20	0.70	2.72	0.54	3.26	−2.56
OAEU$_{L6}$: Same as OAEU, with leaching rate/6	0.40	0.08	0.02	0.20	0.70	0.51	0.59	1.10	−0.40
OAEU$_{L12}$: Same as OAEU, with leaching rate/12	0.40	0.08	0.02	0.20	0.70	0.26	0.60	0.86	−0.16
Belon et al (2012), France	1.02	0.56	0.00	0.25	1.83	n.d.	n.d.	n.d.	n.d.
Six and Smolders (2014), Europe	0.79	0.06	0.09	0.35	1.29	2.56	0.20	2.76	−1.47

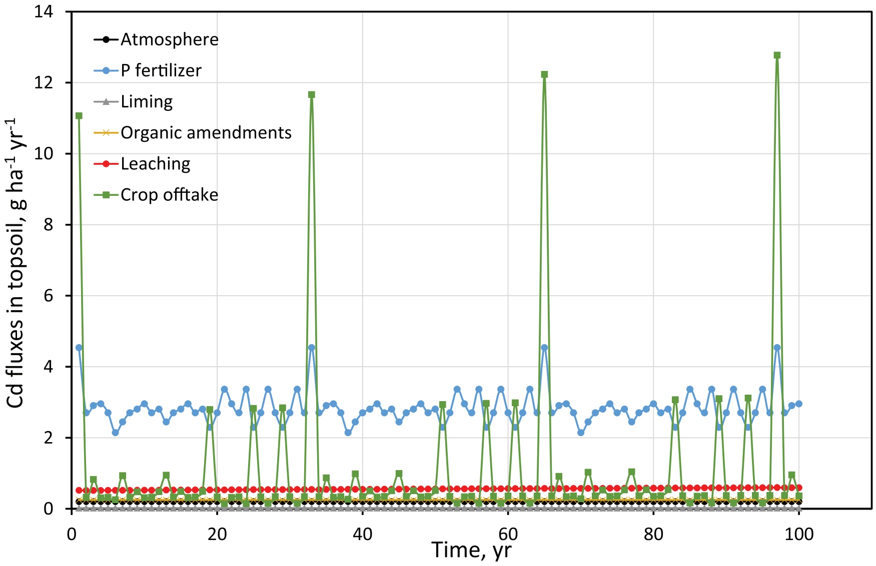

Fig. 3. Estimated annual cadmium fluxes in the ploughed layer of the mean French soil under annual crops for scenario CPA_{L6}.

conventional agriculture), crop offtake decreased to a value around 0.6 g Cd ha^{-1} yr^{-1}, as could be expected from Eq. (13), but also because the soil Cd content decreased and the rotation changed, including less forage maize and sugar beet which export the most Cd (Table 7).

In contrast, the Cd input due to P fertilizer application consistently varied according to the scenario. In conventional agriculture, a reduction in the Cd content in the P fertilizer (EUR scenario) would reduce the average Cd input of 55% compared to the CPA scenario (from 2.84 to 1.27 g Cd ha^{-1} yr^{-1}, Table 6). The application of good practices for P fertilization (GPPA scenario) would have a similar effect to that of the EUR scenario, with a Cd input due to P fertilizer of 1.07 g Cd ha^{-1} yr^{-1}. Fertilizer contribution would be only 0.47 g Cd ha^{-1} yr^{-1} in the GPEU scenarios, combining a reduction in the Cd content in the fertilizer and in the fertilizer application rate. In scenarios of organic agriculture (OA and AOEU), the Cd input through P fertilization was 0.83 g Cd ha^{-1} yr^{-1} and 0.47 g Cd ha^{-1} yr^{-1} respectively, which is a little below those of the corresponding good practice scenarios for conventional agriculture (GPPA and GPEU).

Excluding the scenario with leaching according to Eq. (8), only scenarios GPEU and OAEU (L6 and L12) led to a negative balance, of the order of −0.2 to −0.6 g Cd ha^{-1} yr^{-1}. In contrast, the balance of CPA_{L6} and CPA_{L12} were positive, with an accumulation of 1.65 g Cd ha^{-1} yr^{-1} and 1.92 g Cd ha^{-1} yr^{-1}, respectively (Table 6). These values can be compared to the estimated past Cd accumulation: Considering that the pedo-geochemical background content is 0.10 mg Cd kg^{-1} (Sterckeman et al., 2006), it can be calculated that 735 g Cd ha^{-1} accumulated in the topsoil to reach its current mean Cd content of 0.31 mg Cd kg^{-1}. Hypothesizing that Cd accumulation started 150 years ago, with the general use of P fertilizers and the increase of atmospheric emission which reached its peak in the middle of the 20th Century (Pacyna et al., 2007), the past mean annual balance can be estimated to be of the order of 5 g Cd ha^{-1} yr^{-1}. It therefore seems that the current and future Cd accumulation rate in French soils would be much lower than what it was in the past. This is consistent with what de Vries and McLaughlin (2013) estimated for Australian agricultural systems.

The Cd inputs can also be compared to those recently estimated for France by Belon et al. (2012) (Table 6). The input from atmospheric deposition and liming in our study are close to those of these authors. The inputs from organic amendments (0.25 g Cd ha^{-1} yr^{-1}) are half those of Belon et al. (2012) (0.56 g Cd ha^{-1} yr^{-1}), but in both cases well below the Cd input from P fertilizers. The mean fertilizer input which can be obtained from their work, is 1.02 g Cd ha^{-1} yr^{-1} at the national scale, while it was 2.84 g Cd ha^{-1} yr^{-1} in the CPA scenarios of this study. The difference could be due to the calculation method. Belon et al. (2012) apparently used the same reference as us for the Cd content in P fertilizer (Nziguheba and Smolders, 2008). The Cd input from P fertilizer was calculated by multiplying the P fertilizer delivery (given by the manufacturers association) divided by the UAA. At the national level, the UAA Belon et al. (2012) used was 29,554,440 ha. This includes permanent grassland, orchard and vineyard, which we excluded from the present study (the UAA of annual crops we used was 17,556,765 ha). The P application rates on these permanent crops might be lower than on annual crops. This is in line with the fact that making their calculation at the departmental scale, Belon et al. (2012) observed the highest Cd

Table 7
Mean cadmium input with P fertilizer application and Cd crop offtake for each crop and three contrasting scenarios.

Crop	P fertilizer input				Offtake		
	CPA_{L12}	$GPEU_{L12}$	$OAEU_{L12}$	CPA_{L12}-$GPEU_{L12}$	CPA_{L12}	$GPEU_{L12}$	$OAEU_{L12}$
	g Cd ha^{-1} yr^{-1}			% CPA_{L12}	g Cd ha^{-1} yr^{-1}		
Barley	2.81	0.33	0.23	88.3	0.51	0.47	0.35
Durum wheat	2.91	0.58	0.45	80.0	0.90	0.82	0.62
Field pea			0.49				0.11
Forage maize	2.30	0.52	0.38	77.5	3.00	2.69	2.03
Grain maize	2.96	0.62	0.48	78.9	0.34	0.31	0.23
Rape	3.37	0.69	0.51	79.6	0.16	0.14	0.11
Sugar beet	4.54	1.05	0.82	76.9	12.07	10.97	8.27
Sunflower	2.45	0.03	0.50	98.8	1.00	0.92	0.69
Triticale	2.14	0.22	0.16	89.8	0.27	0.25	0.19
Wheat	2.70	0.40	0.30	85.1	0.35	0.32	0.24

inputs in arable crop regions. However, the Cd flux data at this scale are not available from their work.

When projecting the current practice into the future, the Cd balance that we calculated for France (+1.65 g Cd ha^{-1} yr^{-1} to +1.92 g Cd ha^{-1} yr^{-1}) is rather different to that simulated for the EU (27 + 1) by Six and Smolders (2014), which is clearly negative (−1.47 g Cd ha^{-1} yr^{-1}). This difference is due to the lower Cd input from P fertilizer application at the European level (as already

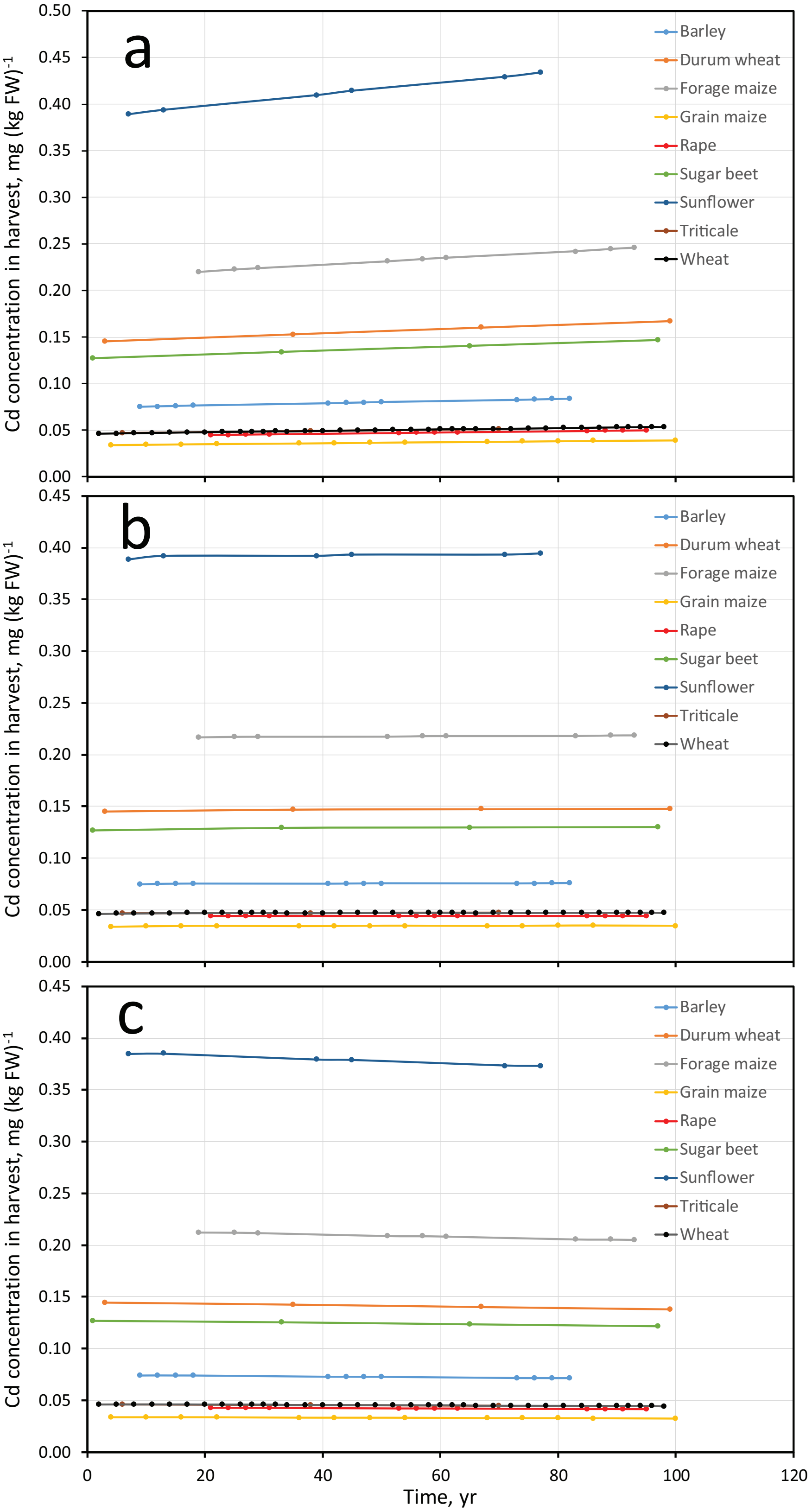

Fig. 4. Evolution of the estimated Cd content in crop harvests for three scenarios. a: CPA_{L6} Scenario. b: EUR_{L6} Scenario. c: $GPEU_{L6}$ Scenario.

mentioned in the Introduction) and to the higher leaching flow (see above).

In some regions, the current Cd input by organic amendments could be reduced if the amount of nitrogen added were more precisely taken into account for the N fertilization calculation. According to simulations of scenarios including fertilization good practice, it is particularly the case in Brittany, were the excess of applied organic amendments (mainly slurry and manure) is 76% more than crop N requirements (Table S6). This also leads to an excess of N and P application to soils, which become available for leaching and coastal eutrophication (Ifremer, 2001). This is also the case in Auvergne and Limousin, where the current excess of organic amendment application is 25% and 20%, respectively.

3.3. Cadmium in crops

At the national level, the crops with the highest Cd inputs are sugar beet and rape (Table 7), which receive the highest P fertilization, while those which most export the metal, are sugar beet, forage maize and sunflower.

Of course, the soil Cd total content whose balance is simulated, is not completely available for plant uptake. In cultivated soils, available Cd is about 50% of total Cd (Sterckeman et al., 2009). In the balance model, the Cd content in the harvested plant part is proportional to the soil Cd total concentration, following Eq. (14), which formalises the soil-to-plant transfer of the metal. Therefore, in the case of the CPA_{L6} scenario, Cd in crops should increase in the future (Fig. 4), contrarily to the recommendation of ANSES (2011) and of the European Commission (Borg, 2014). This should however remain below the content limits fixed by the regulations for foodstuffs (0.2 mg Cd $(kg\ FW)^{-1}$ for wheat, 0.1 mg Cd $(kg\ FW)^{-1}$ for other cereals) (European Commision, 2006) and for animal feed (1 mg Cd $(kg\ FW)^{-1}$) (Geslain-Lanéelle and Gallot, 2001). It should even slightly decrease in the $GPEU_{L6}$ scenario, and be quasi-stationary in the case of the EUR_{L6} scenario.

3.4. How to improve forecasts

Of course, these forecasts are based on simulations which suffer from many uncertainties. The accuracy of all the balance items could be ameliorated, but a priority should be given to those of the soil Cd outputs. Indeed, the estimated Cd inputs from fertilizer (applied dose, Cd concentration in fertilizer) are based on strong statistical data. If the inputs from the amendments might not be as good, a greater precision at this level would not consistently change the balances, as the various amendments constitute a minor part of the Cd inputs, at least as far as the contribution of P fertilizers is so dominant.

Among the Cd outputs, the contribution of the crop has the merit of being based on numerous measurements, although the transfer factor is a simplistic way of representing the soil-to-plant transfer of Cd. A crop model formalizing the root uptake according to the soil metal availability and the distribution of Cd in the various plant organ would be a means of improving the prediction of Cd crop offtake. The other output item, the estimation of Cd leaching out of the ploughed horizon is more critical, since it is based on a too simplistic model, which has not been experimentally validated. On the contrary, the few field measurements available, show that Cd leaching is highly overestimated. What's more, Cd leaching in poorly contaminated soils is very difficult to measure or model. To date, there are very few data about Cd concentration in percolating water and about Cd leaching under annual crops, in France or in the other parts of the world. To fill this gap, measurements should be carried out under various agro-environmental contexts and efforts should also be made to develop a better leaching model, whose formalism should be adapted to the data available at the scale at which the balances are simulated.

The predictions of the model give average values which can be considered to control the chronic exposure of the population as a whole. However, they could be ameliorated, by associating an uncertainty analysis to characterise the distribution of the model results, using a Monte Carlo approach (Saltelli et al., 2004). Further investigations would then be necessary to define the distribution of each of the numerous input variables.

4. Conclusions

If current cultivation practices are continued, the average Cd content in French soils under annual crops will increase by about 15% by the end of the next century. The concentration of the metal in the crops would increase in the same proportions, thereby intensifying the chronic Cd exposure of human populations through their dietary intake. The cause of this increase is the input of Cd with P fertilizer applications, which represents around 85% of Cd inputs in soil, and which is nearly twice the Cd outputs by leaching and crop offtake.

These results concerning French soils are opposite to those recently obtained at the European level, which concluded that there would be an average decrease of 15% in the soil Cd content. The main reasons of this contradiction are 1) a higher rate of P application in France than in Europe, 2) a higher Cd content in the French P fertilizers compared to the European ones and 3) lower Cd leaching in French soils, the leaching rate calculated in the study at the European level probably being overestimated.

In France, P applications on annual crops are excessive and could be reduced by about 50%, while still satisfying the crop requirements. Therefore, strictly applying the good practices for P fertilization as recommended by COMIFER would stabilize the future soil Cd content at its present level. Assuming current excessive P fertilization, the enforcement of a regulation limiting Cd content in P fertilizers, as by proposed the European Union, would lead to a clearly lesser increase in soil Cd, of between 1.6% and 3.9% after 100 years. Only the combination of P fertilization good practices and of a regulation limiting Cd in P fertilizers would lead to a decrease of Cd in soil, of between 3.0% and 5.2%, after a century of conventional agriculture.

Organic agriculture would lead to an evolution of soil Cd content similar to that of conventional agriculture applying good practices for P fertilization. That means that the soil Cd content would not or would only slightly increase in a scenario with a total conversion to organic agriculture, and decrease if Cd content in phosphate rock used as the only P source is reduced.

Acknowledgements

This work was supported by the Ministère de l'Agriculture, de l'Agroalimentaire et de la Forêt, within the framework of the VARHYPEX programme (convention AAP no C-2015-12). We are grateful to the anonymous people from the AGRESTE department of the same Ministry, who provided us with data and complementary information. We also thank Sylvie Dauguet from Terres Inovia and Hervé Escriou, from ITB, as well as Sabine Houot and Nicolas Beaudouin from INRA, for providing various data. The authors are grateful to Denis Montenach (INRA, Colmar) for access to the PRO'spective field experiment, which forms part of the SOERE-PRO (network of long-term experiments dedicated to the study of impacts of organic waste product recycling) certified by ALLENVI (Alliance Nationale de Recherche pour l'Environnement) and integrated as a department of the “Investment d'Avenir” infrastructure AnaEE-France, overseen by the French National Research Agency (ANR-11-INBS-0001). ThePRO'spective field experiment is conducted by INRA Colmar in collaboration with SMRA68 (Syndicat Mixte Recyclage Agricole du Haut-Rhin), ARAA (Association pour la Relance Agronomique en Alsace) and UHA (Université de Haute-Alsace). The field experiment is financed by SMRA68, ADEME, AERM, Veolia, SITEUCE, Arvalis, Terralys, SEDE, COVED, SM4.

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.05.225.

References

AGRESTE, 2014. Agreste les Dossiers no 21 - Enquête Pratiques Culturales 2011. Ministère de l'Agriculture, de l'Agroalimentaire et de la Forêt, Paris.

ANSES, 2011. Étude de l'alimentation totale française 2 (EAT 2). Tome 1. Contaminants Inorganiques, Minéraux, Polluants Organiques Persistants, Mycotoxines, Phyto-estrogènes. ANSES, Maison-Alfort, p. 346.

ARVALIS, 2016. Plan de fumure. Intégrer les valeurs fertilisantes des produits organiques. ARVALIS - Institut du Végétal, Paris https://www.arvalis-infos.fr/view-12357-arvarticle.html.

ATSDR, 2012. Toxicological Profile for Cadmium. U.S. Department of Health and Human Services, Public Health Service, Agency for Toxic Substances and Disease Registry, Atlanta, p. 487.

Baize, D., Deslais, W., Gaiffe, M., 1999. Anomalies naturelles en cadmium dans les sols de France. Etude et Gestion des Sols. 6 pp. 85–104.

Bech, J., Suarez, M., Reverter, F., Tume, P., Sánchez, P., Roca, N., et al., 2010. Selenium and other trace element in phosphorites: a comparison between those of the Bayovar-Sechura and other provenances. J. Geochem. Explor. 107, 146–160.

Belon, E., Boisson, M., Deportes, I.Z., Eglin, T.K., Feix, I., Bispo, A.O., et al., 2012. An inventory of trace elements inputs to French agricultural soils. Sci. Total Environ. 439, 87–95.

Bengtsson, H., Alvenäs, G., Nilsson, S.I., Hultman, B., Öborn, I., 2006. Cadmium, copper and zinc leaching and surface run-off losses at the Öjebyn farm in Northern Sweden—temporal and spatial variation. Agric. Ecosyst. Environ. 113, 120–138.

Blume, H.-P., Brümmer, G.W., Fleige, H., Horn, R., Kandeler, E., Kögel-Knabner, I., et al., 2016. Chemical Properties and Processes. Scheffer/SchachtschabelSoil Science. Springer Berlin Heidelberg, Berlin, Heidelberg, pp. 123–174.

Borg, T., 2014. Recommandation de la Commission du 4 avril 2014 sur la réduction de la présence de cadmium dans les denrées alimentaires. In: Européenne C (Ed.), 2014/193/UE. Commission Européenne, Bruxelles L104/80-L104/81.

Bruand, A., Duval, O., Cousin, I., 2004. Estimation des propriétés de rétention en eau des sols à partir de la base de données SOLHYDRO: une première proposition combinant le type d'horizon, sa texture et sa densité apparente. Etude et Gestion des Sols. 11 pp. 323–334.

Cambier, P., Pot, V., Mercier, V., Michaud, A., Benoit, P., Revallier, A., et al., 2014. Impact of long-term organic residue recycling in agriculture on soil solution composition and trace metal leaching in soils. Sci. Total Environ. 499, 560–573.

Carpentier, M., 2014. Coût de production du blé tendre. A 200 €/t selon Arvalis, «un niveau trop élevé par rapport au marché». Terre-net. MEDIA DATA SERVICES, Beauvais.

Clemens, S., Aarts, M.G.M., Thomine, S., Verbruggen, N., 2013. Plant science: the key to preventing slow cadmium poisoning. Trends Plant Sci. 18, 92–99.

COMIFER, 1995. Aide au diagnostic et à la prescription de la fertilisation phosphatée et potassique des grandes cultures. COMIFER, Paris.

COMIFER, 2009. Fertilisation PK. Grille de calcul de dose, Paris.

COMIFER, 2011. Calcul de la fertilisation azotée. Guide méthodologique pour l'établissement des prescriptions locales. Cultures annuelles et prairies. Paris La Défense, COMIFER.

COMIFER, 2016. Objectif, domaine de validité et procédure d'utilisation de la calculette permettant de calculer Mh. Notice d'utilisation (Version 2).

Degryse, F., Smolders, E., 2006. Mobility of Cd and Zn in polluted and unpolluted Spodosols. Eur. J. Soil Sci. 57, 122–133.

DG Enterprise, 2003. Proposal for a Regulation of the European Parliament and of the Council relating to cadmium in fertilizers. Extended impact assessment. Draft No22 of 31.07.2003. European Commission.

Duigou, N., Baize, D., 2010. Nouvelle collecte nationale d'analyses d'éléments en traces dans les sols (horizons de surface) – (Cd, Cr, Cu, Hg, Ni, Pb, Se, Zn). ADEME.

EFSA, 2012. Cadmium dietary exposure in the European population. EFSA J. 10, 2551.

European Commisison, 2006. Commission Regulation (EC) No1881/2006 of 19 December 2006 Setting Maximum Levels for Certain Contaminants in Foodstuffs. European Union L364/5-24.

European Commisison, 2008. In: Union_Européenne (Ed.), COMMISSION REGULATION (EC) No 889/2008 of 5 September 2008 Laying Down Detailed Rules for the Implementation of Council Regulation (EC) No 834/2007 on Organic Production and Labelling of Organic Products with Regard to Organic Production, Labelling and Control, p. 84.

European Commisison, 2016. Proposal for a regulation of the European Parliament and of the Council laying down rules on the making available on the market of CE marked fertilising products and amending Regulations (EC) No 1069/2009 and (EC) No 1107/2009. In: Commission E (Ed.), COM(2016) 157 Final. European Commission, Brussels, p. 43.

European Parliament, 2017. CE Marked Fertilising Products ***I. Amendments Adopted by the European Parliament on 24 October 2017 on the Proposal for a Regulation of the European Parliament and of the Council Laying Down Rules on the Making Available on the Market of CE Marked Fertilising Products and Amending REGULATIONS (EC) No 1069/2009 and (EC) No 1107/2009 (COM(2016)0157 – C8–0123/2016–2016/0084(COD)). In: Parliament E, editor. P8_TA-PROV(2017)0392. European Parliament, Strasbourg, p. 116.

Eurostat, 2013. Agriculture, Forestry and Fishery Statistics: European Union.

Eurostat, 2017. Organic crop area continues to rise in the EU. European Commission, Brussels http://ec.europa.eu/eurostat/web/products-eurostat-news/-/DDN-20171116-1?inheritRedirect=true&redirect=%2Feurostat%2Fweb%2Fmain%2Fhome.

Fertilizers Europe, 2014. Decadmiation workshop. Putting All the Cards on the Table. Fertilizer International. BCInsight Ltd, London, pp. 28–31.

Filipović, V., Cambier, P., Filipović, L., Coquet, Y., Pot, V., Bodineau, G., et al., 2016. Modeling copper and cadmium mobility in an Albeluvisol amended with urban waste composts. Vadose Zone J. 15, 1–15.

FranceAgriMer, 2015. L'observatoire national des ressources en biomasse. Évaluation des ressources disponibles en France. LES SYNTHÈSES de FranceAgriMer, Montreuil-sous-Bois. 114.

Geslain-Lanéelle, C., Gallot, J., 2001. Arrêté du 12 janvier 2001 modifiant l'arrêté du 16 mars 1989 fixant les teneurs maximales pour les substances et produits indésirables dans l'alimentation des animaux. J. Officiel de la République Française 1049–1055 République Française, France.

Habets, F., Boone, A., Champeaux, J.L., Etchevers, P., Franchistéguy, L., Leblois, E., et al., 2008. The SAFRAN-ISBA-MODCOU hydrometeorological model applied over France. J. Geophys. Res.-Atmos. 113 (1984–2012).

Houot, S., Pons, M.-N., Pradel, M., Tibi, A., Aubry, C., Augusto, L., et al., 2014. Valorisation des matières fertilisantes d'origine résiduaire sur les sols à usage agricole ou forestier. Impacts Agronomiques, Environnementaux, Socio-économiques. Expertise Scientifique Collective, Paris, p. 930.

Ifremer, Ménesguen, A. (Eds.), 2001. L'eutrophisation des eaux marines et saumâtres en Europe, en particulier en France. Ifremer, Plouzané (France).

Ilyin, I., Rozovskaya, O., Travnikov, O., Varygina, M., Aas, W., Pfaffhuber, K.A., 2016. Assessment of heavy metal transboundary pollution, progress in model development and mercury research. EMEP Status Report 2/2016. Meteorological Synthesizing Centre - East, Norwegian Institute for Air Research (NILU), p. 84.

Järup, L., Åkesson, A., 2009. Current status of cadmium as an environmental health problem. Toxicol. Appl. Pharmacol. 238, 201–208.

Khasawneh, F.E., Doll, E.C., 1979. The use of phosphate rock for direct application to soils. In: Brady, N.C. (Ed.), Advances in Agronomy. 30. Academic Press, pp. 159–206.

Leifeld, J., Fuhrer, J., 2010. Organic farming and soil carbon sequestration: what do we really know about the benefits? Ambio 39, 585–599.

Lopez, J., 2002. Les composts. Le Courrier de l'Environnement de l. INRA, Paris.

Mclaughlin, M.J., Tiller, K.G., Naidu, R., Stevens, D.P., 1996. Review: the behaviour and environmental impact of contaminants in fertilizers. Aust. J. Soil Res. 34, 1–54.

Muller, A., Schader, C., El-Hage Scialabba, N., Brüggemann, J., Isensee, A., Erb, K.-H., et al., 2017. Strategies for feeding the world more sustainably with organic agriculture. Nat. Commun. 8, 1290.

Nziguheba, G., Smolders, E., 2008. Inputs of trace elements in agricultural soils via phosphate fertilizers in European countries. Sci. Total Environ. 390, 53–57.

Pacyna, E.G., Pacyna, J.M., Fudala, J., Strzelecka-Jastrzab, E., Hlawiczka, S., Panasiuk, D., et al., 2007. Current and future emissions of selected heavy metals to the atmosphere from anthropogenic sources in Europe. Atmos. Environ. 41, 8557–8566.

Quezada-Hinojosa, R.P., Matera, V., Adatte, T., Rambeau, C., Föllmi, K.B., 2009. Cadmium distribution in soils covering Jurassic oolitic limestone with high Cd contents in the Swiss Jura. Geoderma 150, 287–301.

Saby, N.P.A., Lemercier, B., Arrouays, D., Leménager, S., Louis, B.P., Millet, F., et al., 2014. Le programme Base de Données des Analyses de Terre (BDAT): Bilan de 20 ans de collecte de résultats d'analyses. Etude et Gestion des Sols. 21, pp. 25–35.

Saltelli, A., Tarantola, S., Campolongo, F., Ratto, M., 2004. Sensitivity analysis in practice. A Guide to Assessing Scientific Models. John Wiley & Sons, Chichester.

Selim, H.M., Elbana, T.A., Zhao, K., Xu, J., Fergusson, E.L., 2013. Miscible displacement of zinc in soil columns: linear and nonlinear modeling. Soil Sci. Soc. Am. J. 77, 391–402.

Six, L., Smolders, E., 2014. Future trends in soil cadmium concentration under current cadmium fluxes to European agricultural soils. Sci. Total Environ. 485–486, 319–328.

Sterckeman, T., Douay, F., Baize, D., Fourrier, H., Proix, N., Schvartz, C., 2006. Trace elements in soils developed in sedimentary materials from Northern France. Geoderma 136, 912–929.

Sterckeman, T., Carignan, J., Srayeddin, I., Baize, D., Cloquet, C., 2009. Availability of soil cadmium using stable and radioactive isotope dilution. Geoderma 153, 372–378.

UNIFA, 2014. Evolution des bilans régionaux de fertilisation en France de 1988 à 2013 pour les trois éléments nutritifs: Phosphore, potassium, magnésium. Union des industries de la fertilisation (UNIFA), La Défense. p. 42.

Union Européenne, 2016. Proposition de Règlement du Parlement Européen et du Conseil établissant les règles relatives à la mise à disposition sur le marché des fertilisants porteurs du marquage CE et modifiant les règlements (CE)n° 1069/2009 et (CE) n° 1107/2009. In: Européenne U, editor. 2016/0084 (COD). Ares(2016)1341463, Commission Européenne. p. 45.

de Vries, W., McLaughlin, M.J., 2013. Modeling the cadmium balance in Australian agricultural systems in view of potential impacts on food and water quality. Sci. Total Environ. 461–462, 240–257.

de Vries, W., Mclaughlin, M.J., Groenenberg, J.E., 2011. Transfer functions for solid–solution partitioning of cadmium for Australian soils. Environ. Pollut. 159, 3583–3594.

Wopereis, M.C., Gascuel-Odoux, C., Bourrie, G., Soignet, G., 1988. Spatial variability of heavy metals in soil on a one-hectare scale. Soil Sci. 146, 113–118.

SUPPLEMENTARY MATERIAL

to

Cadmium balance in French soils under annual crops: Scenarios for the next century

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1. Supplementary data concerning the materials and methods

1.1. UAA

Table S1. Utilised agricultural area (UAA) of the different spatial units, provided by AGRESTE, after an enquiry in 2010.

Spatial unit	Annual crops	Permanent grassland	Total
Alsace	229598	81621	311219
Aquitaine	793364	408341	1201705
Auvergne	556875	917276	1474151
Bourgogne	980617	807405	1788022
Bretagne	1508124	206533	1714657
Centre	1917601	303727	2221328
Champagne-Ardenne	1187983	275820	1463803
Corse	9434	385670	395104
Franche-Comté	279454	424166	703620
Ile de France	518829	30561	549390
Languedoc-Roussillon	217428	448538	665966
Limousin	291576	567500	859076
Lorraine	694581	440000	1134581
Midi-Pyrénées	1520794	853009	2373803
Nord-Pas de Calais	617349	179675	797024
Basse-Normandie	672334	599354	1271688
Haute-Normandie	585682	203509	789191
Pays de Loire	1688118	512990	2201108
Picardie	1128221	168708	1296929
Poitou-Charentes	1389634	191553	1581187
Provence-Alpes	165343	470062	635405
Rhône-Alpes	603826	855733	1459559
France	17556765	9331751	26888516

1.2. Data concerning P fertilization

Table S2. Average P fertilizer application rates in France and in its 22 regions, for different crops (AGRESTE, 2014). The P content in the harvested plant part (P_{har}) is also given, as well as the C_r coefficient (COMIFER, 2009), both used for the calculation of the P fertilization dose according to good practices (COMIFER, 1995).

Crop	P content of harvested part P_{har} kg P_2O_5 (t FW)$^{-1}$	C_r	P fertilizer application, for each spatial unit (kg P_2O_5 ha^{-1})											
			France	Alsace	Aquitaine	Auvergne	Basse-Normandie	Bourgogne	Bretagne	Centre	Champagne-Ardenne	Corse	Franche-Comté	Haute-Normandie
Barley	6.5	1	30			32	23	36	25**	37	38	30^N	30	22
Durum wheat	8.5	1	28							26				
Field pea	8.0	1	31				31	31	26**	36	52			23
Forage maize	4.2	1	25	40***	46	21	34	23	21	15	33***	25^N	40	33
Grain maize	6.0	1	41	66	53	46	31	52	15	46	54	41^N	37	
Potato	1.0	1.5	53				38		54	102	89			53
Rape	12.5	1.5	37			37	28	38	31**	42	53		30	31
Sugar beet	0.5	1.5	47	75***			54			74	77			40
Sunflower	12.0	1	24		32	35		35		26	54		$53^{\#}$	
Triticale	6.5	1	14		35	19	10	19	12**	25		14^N	17	
Wheat	6.5	1	20	23	40	24	12	34	17**	32	35	20^N	44	16

Crop	P fertilizer application, for each spatial unit (kg P_2O_5 ha^{-1})										
	Île-de-France	Languedoc-Roussillon	Limousin	Lorraine	Midi-Pyrénées	Nord-Pas-de-Calais	Pays de la Loire	Picardie	Poitou-Charentes	Provence-Alpes-Côte d'Azur	Rhône-Alpes
Barley	22	$33^{\$}$	17**	30	13	14	$13^{\#}$	30	30	$20^{\$\$}$	29***
Durum wheat		$31^{\$}$			44				36	19	31
Field pea	34			23	22		15	28	20		
Forage maize			14	27	34***	38	20	24	24	$17^{\$\$}$	27
Grain maize	44		23**	40	56	24	18	32	33	$28^{\$\$}$	40
Potato	44					48		35			
Rape	31	$40^{\$}$		30	42	17	$16^{\#}$	25	49	$25^{\$\$}$	36***
Sugar beet	29					37		31			
Sunflower		26			28		12		15	$16^{\$\$}$	30
Triticale		11	25	21	12		6		10	$10^{\$\$}$	20
Wheat	10	$22^{\$}$	11**	28	31	20^N	8	7	12	$13^{\$\$}$	23

**estimated from the ratio of regional to national application rates for forage maize

***estimated from the ratio of regional to national application rates for grain maize

$^{\#}$estimated from the ratio of regional to national application rates for wheat

$^{\$}$estimated from the ratio of regional to national application rates for sunflower

$^{\$\$}$estimated from the ratio of regional to national application rates for durum wheat

^{N}national mean

Table S3. Average crop yields (t ha^{-1}) in France and in its 22 regions; mean values calculated from 2011 to 2015, with data provided by AGRESTE (https://stats.agriculture.gouv.fr/disar/).

Crop	**France**	**Alsace**	**Aquitaine**	**Auvergne**	**Basse-Normandie**	**Bourgogne**	**Bretagne**	**Centre**	**Champagne-Ardenne**	**Corse**	**Franche-Comté**	**Haute-Normandie**
Barley	6.4	6.2	5.5	5.4	7.1	5.9	7.0	6.8	6.8	2.9	5.9	7.9
Durum wheat	5.7		5.0	5.0	5.2	5.3		6.6	5.4	3.5		5.8
Field pea	3.7	3.4	2.3	3.0	4.4	3.4	4.5	3.7	4.1	2.3	3.3	4.6
Forage maize	12.7	14.4*	13.9*	10.8*	14.0*	10.7*	13.5*		11.6*	9.9*	12.3*	14.6*
Grain maize	9.1	11.1	9.2	9.4	8.6	8.9	8.8	9.6	8.6	10.8	8.8	8.7
Potato	35.6	41.1	24.4	29.5	29.3	35.5	31.2	49.4	51.1	25.0	29.4	44.2
Rape	3.3	3.7	2.6	2.8	3.7	3.1	3.5	3.3	3.6		3.5	3.9
Sugar beet	87.1	89.6	82.5	87.0	87.5	84.0	78.3	94.0	91.9			90.7
Sunflower	2.4	2.6	2.4	2.6	2.6	2.3	2.3	2.4	2.5	1.8	2.3	2.8
Triticale	5.5	5.5	5.0	5.0	5.9	4.6	6.5	5.2	6.0	3.4	5.1	5.7
Wheat	7.0	7.4	5.7	6.1	7.6	6.5	7.4	7.2	8.1	3.4	6.5	8.8

Crop	**Île-de-France**	**Languedoc-Roussillon**	**Limousin**	**Lorraine**	**Midi-Pyrénées**	**Nord-Pas-de-Calais**	**Pays de la Loire**	**Picardie**	**Poitou-Charentes**	**Provence-Alpes-Côte d'Azur**	**Région Rhône-Alpes**
Barley	7.3	4.1	5.3	6.0	4.8	8.3	6.5	7.7	5.9	3.9	5.4
Durum wheat	6.6	3.9		5.0	5.1	6.0	6.7	6.0	6.1	3.6	4.9
Field pea	4.3	2.7	3.0	3.7	3.1	5.0	3.9	4.4	3.5	2.3	3.3
Forage maize			12.5*	11.3*	12.5*	15.0*	12.7*	14.8*	11.4*		10.9*
Grain maize	9.9	8.8	7.5	8.0	9.8	9.8	8.9	9.4	8.9	11.0	9.6
Potato	46.2	23.4	26.0	40.9	28.1	46.2	30.8	46.5	20.4	28.6	23.9
Rape	3.7	2.7	2.9	3.2	2.7	4.2	3.3	4.0	3.2	1.9	3.1
Sugar beet	88.8			87.6		90.5	83.4	87.7			
Sunflower	2.9	2.0	2.0	2.5	2.1		2.5	2.3	2.2	1.8	2.3
Triticale	6.3	3.7	5.1	5.6	4.4	7.1	5.9	6.3	5.0	3.9	5.2
Wheat	8.3	4.6	5.5	6.7	5.4	9.0	7.1	8.8	6.5	3.9	6.0

* t DM yr^{-1}

Table S4. Threshold values for the diagnosis of the P_2O_5 Olsen content in soils. Mean values were calculated for each region and for France as a whole, using the data from ARVALIS (2009). In absence of values for some regions, national values were considered.

Spatial unit	**High P requirement crop**		**Middle P requirement crop**		**Low P requirement crop**	
	Trenf	**Timp**	**Trenf**	**Timp**	**Trenf**	**Timp**
Alsace	56	86	56	86	26	76
Aquitaine	40	80	30	80	20	45
Auvergne	52	82	52	82	22	72
Bourgogne	53	83	53	83	23	73
Bretagne	54	84	54	84	24	74
Centre	54	84	54	84	24	74
Champagne-Ardenne	69	105	67	95	30	84
Corse						
Franche-Comté	51	81	51	81	21	71
Ile de France	54	84	54	84	24	74
Languedoc-Roussillon						
Limousin	53	83	53	83	23	73
Lorraine	54	84	54	84	24	74
Midi-Pyrénées	40	80	30	80	20	45
Nord-Pas de Calais	74	113	70	98	30	86
Basse-Normandie	53	83	53	83	23	73
Haute-Normandie	53	83	53	83	23	73
Pays de Loire	54	84	54	84	24	74
Picardie	74	113	70	98	30	86
Poitou-Charentes	65	99	63	92	29	82
Provence-Alpes-Côte d'Azur						
Rhône-Alpes	53	83	53	83	23	73
France	55	88	53	85	24	72

1.3. Composition of the organic amendments

Table S5. Composition of the organic amendments. N, P_2O_5 and Cd contents were obtained from Houot et al. (2014). K_{eq} was taken from internet publications by ARVALIS – Institut du Végétal (ARVALIS, 2011; ARVALIS, 2016).

Amendment	**Composition basis**	**N**		**P_2O_5**		**Cd**		**DM**
		g kg^{-1}	**K_{eq}**	**g kg^{-1}**	**K_{eq}**	**mg kg^{-1}**	**K_{Cd}**	**%**
Manure	FW	5.7	0.2	3	0.8	0.1	0.18	35
Slurry	FW	4.5	0.5	2.5	0.95	0.01	1	3.63
Urban compost	FW	8.3	0.1	4.2	0.55	0.3	1	59
Sludge	DM	1.6	0.2	83	0.9	1.4	1	
Industrial organic wastes	DM	63.4	0.2	49.2	0.9	1	1	
Sludge and effluents from agri-food industry	DM	63.4	0.2	49.2	0.9	1	1	
Sludge and effluents from other industries	DM	24.9	0.2	10.5	0.9	1	1	

Table S6. Amendment quantities spread on soils under annual crops in each French region.

Spatial unit	Manure	Slurry	Urban compost	Sludge	Industrial organic wastes	Sludge and effluents from agri-food industry	Sludge and effluents from other industries	Total, scenarios CPA et EUR	Total, scenarios GPPA et GPEU	Present amendment excess in confront to N fertilization needs
	kg DM ha^{-1} yr^{-1}							kg DM ha^{-1} (100 yrs)$^{-1}$		%
Alsace	1008	166	110	55	5.1	22.1	8.5	137464	137464	0
Aquitaine	1091	195	91	28	21.2	92.0	35.4	155288	155288	0
Auvergne	1390	346	16	9	6.7	29.1	11.2	180808	144647	25
Bourgogne	1039	204	23	7	5.0	21.8	8.4	130811	130811	0
Bretagne	2112	562	90	21	32.1	139.0	53.5	300945	170896	76
Centre	493	79	32	17	0.8	3.5	1.4	62698	62698	0
Champagne-Ardenne	729	105	17	12	11.5	49.7	19.1	94372	94372	0
Corse	168	82	6	3	0.0	0.0	0.0	26024	26024	0
Franche-Comté	1566	262	38	23	1.7	7.4	2.8	190162	182556	4
Île de France	96	21	219	187	4.4	18.9	7.3	55365	55365	0
Languedoc-Roussillon	398	126	91	53	0.3	1.4	0.5	67078	67078	0
Limousin	1460	385	17	6	0.0	0.0	0.0	186728	156214	20
Lorraine	1386	225	31	27	7.1	30.9	11.9	171980	171980	0
Midi-Pyrénées	918	165	26	10	2.0	8.8	3.4	113363	113363	0
Nord-Pas de Calais	1507	195	142	70	36.1	156.6	60.2	216759	216759	0
Basse-Normandie	1747	429	43	20	2.8	12.3	4.7	225842	225842	0
Haute-Normandie	1233	244	94	38	19.8	85.7	32.9	174676	174676	0
Pays de Loire	1747	381	51	19	14.7	63.8	24.5	230046	223256	3
Picardie	698	169	47	11	11.1	48.1	18.5	100221	100221	0
Poitou-Charentes	1033	137	37	16	6.3	27.1	10.4	126613	126613	0
Provence-Alpes-Côte d'Azur	373	76	146	60	14.2	61.4	23.6	75333	75333	0
Rhône-Alpes	1185	230	90	27	4.3	18.7	7.2	156184	144606	8

1.4. Nitrogen requirements

In scenarios where P fertilisation was calculated according to good practices, the amount of N required by the crop was also calculated based on the nitrogen balance method proposed by COMIFER (2011) (http://www.comifer.asso.fr/index.php/fr/bilan-azote.html). As a consequence, when N added by the organic amendment exceeded the crop requirement, the quantity of amendments applied was reduced to balance the N supply with the crop requirement and to reduce N leaching. This was operated through R_N calculated as the ratio of N crop requirement (N_{CR}, kg N ha^{-1}) to N provided by the organic amendments (if $R_N \geq 1$, $R_N = 1$).

The nitrogen balance was simplified to calculate N_{CR} as follows:

$$N_{CR} = (Pf + L + Rf) - (Pi + Mh + Mr + Ri + A)$$

Pf is the quantity of N absorbed by the crop at the end of cultivation (closure of the N balance). For most of the crop, this was calculated multiplying the yield by the N content of the harvested part. L is the amount of leached N; this was taken to be 20 kg N ha^{-1}. Rf is the mineral N which remains in the soil at harvest; this was taken to be 40 kg N ha^{-1}. Pi is the amount of N already absorbed by the crop at the opening of the balance. This value varies according to the crop in question. Mh is the amount provided by mineralization of the organic matter. This was calculated with the Mh model provided by COMIFER (2016) and depends on the climate, soil properties and the cultivation duration. Mr is the amount of N provided by mineralisation of the previous crop residues. It was taken to be 10 kg N ha^{-1}. Ri is the mineral N in the soil at the opening of the balance and its value was taken to be 40 kg N ha^{-1}. A is the amount brought by atmospheric fallout and was taken to be 5 kg N ha^{-1}.

The opening and closure date of the N balance depended on the crop and on the region.

1.5. Cadmium in rock phosphate

We put forward the hypothesis that the rock phosphate would come from Western Sahara, where the biggest phosphate reserves are located (Gilbert, 2009) and are the closest to Europe. Bech et al. (2010) analysed 11 phosphorite samples from this area and. These data, together with those of McLaughlin et al. (1996) lead to a mean content of 28.9 mg Cd kg^{-1} i.e. 87 mg Cd (kg P_2O_5)$^{-1}$ (Table S7). If the regulation n°889/(2008) from the European Commission is respected, rock phosphate applied in organic agriculture should contain a maximum of 90 mg Cd (kg P_2O_5)$^{-1}$. When considering only those samples respecting this condition, the mean content of rock phosphates becomes 47.3 mg Cd (kg P_2O_5)$^{-1}$.

Table S7. Cadmium and phosphorus content in rock phosphate samples.

Country	**Sample**	**Cd mg kg^{-1}**	**P^a %wt**	**P_2O_5 %wt**	**Cd mg (kg P_2O_5)$^{-1}$**
Morocco	Khouribga KIID	22		*32.1*	68.6
Morocco	Khouribga KIISB	17		*32.1*	53.0
Morocco	Khouribga KIISL	16		*32.1*	49.9
Morocco	Khouribga KIIC	17		*32.1*	53.0
Morocco^a	Khouribga 68 K11	12	13.6	31.1	38.5
Morocco^a	Khouribga 72 K20	24	14.4	33.0	72.8
Morocco	Boucraa BGA	3		*33.1*	9.1
Morocco	Boucraa BGB	4		*33.1*	12.1
Morocco	Boucraa BGC	4		*33.1*	12.1
Morocco^a	Boucraa	38	15.7	36.0	105.7
Morocco	Youssoufia YN	63		*32.4*	194.4
Morocco^a	Youssoufia 68 (Y1)	23	13.6	31.1	73.8
Morocco^a	Youssoufia 73 5 Y2	33	14.7	33.7	98.0
Tunisia	Gafsa	53		*33.1*	160.1
Tunisia^a	Gafsa	38	13.4	30.7	123.8
Algeria	Undifferentiated	25.5		*33.1*	77.0
Senegal	Tobene	37.1		*33.1*	112.0
Senegal^a	Undifferentiated	90.0	15.8	36.2	248.7
Mean	All samples	28.9	14.5	33.1	86.8
	< 90 mg Cd (kg P_2O_5)$^{-1}$	15.2			47.3

^{a}From McLaughlin et al. (1996), the other data being from Bech et al. (2010)

1.6. Cadmium imported through manure

Manure is made of straw and cattle excrement. For Cd input through manure, we considered that the metal contained in the straw had previously been exported by this straw from the soil where the manure was spread. Indeed, the straw used for cattle litters was produced in the same spatial unit (region or whole France) as that where the manure was spread. The amount of metal brought to soil by the manure straw was therefore cancelled out by multiplying the amount contained in manure by a $K_{Cd_i} < 1$coefficient. K_{Cd_i} was calculated as follows. According to

AGRESTE (2011), the average use of straw per animal in 2008 was 760 kg DM for a cow. According to Loyon (2015), a cow produces an average of about 2.65 t of fresh manure per year (51.5 10^6 t fresh manure / 19.4 10^6 head of cattle) or 0.93 t of dry manure (considering manure contains 35% of dry matter). The proportion of straw in the manure is therefore 760/930 = 0.82. So 82% of the Cd brought by the manure can be considered as a return of what had been removed with the straw and consequently $K_{Cd_i} = 0.18$.

1.7. Basic amendments

ANPEA (National Professional Association for Fertilizers and Amendments) provided the deliveries of basic mineral amendments in France (Table S8), on a national scale. Regional values were not available.

Table S8. Deliveries of basic mineral amendments in metropolitan France (10^3 t) according to ANPEA (http://www.anpea.com/livraisons-de-fertilisants/livraisons-d-amendements.html).

Year	2011/2012	2012/2013	2013/2014	2014/2015
Lime	213,7	193,7	175	169
Carbonates	1 541,6	1 433,1	1 434	1 323
Mixed amendments	231,3	228,1	220	176
Fertilizing amendments	80,4	68,7	56	67
Other amendments	1 131,6	1 033	1 053	1 144
Total	3 198,5	2 956,9	2 938	2 879
Total CaO from "Lime" and "Carbonates"	*1025*	*949*	*935*	*869*

We took into account only lime (as $Ca(OH)_2$) and carbonates (as $CaCO_3$) as these are the most frequently applied basic amendments on annual crops, but also because the composition of the "mixed amendments", "fertilizing amendments" and "other amendments" were not available. The mean delivery becomes 945 000 t CaO yr^{-1} (mean of the values in the last row of Table S8).

1.8. Evaluation of the leaching modelling

1.8.1. Data from a long term field trial in Sweden

Bengtsson et al. (2006) measured Cd concentrations and flows in percolating soil water and surface run-off on arable land, in an experimental farm from Northern Sweden. They collected soil water for five years, from four sampling sites differing in soil type and agricultural practices. There were three to four replicated water sampling devices (suction cup lysimeters), collecting water at three depths (20, 50 and 80 cm). We compared the Cd concentration measured in the soil solution to that estimated with the model used by Six and Smolders (2014) (Table S9)

Table S9. Comparison of simulated Cd concentrations in percolating water to values measured in the long-term field trial of Öjebyn (Sweden) (Bengtsson et al., 2006).

Plot	Depth cm	pH water	Total C %	Total Cd mg kg^{-1}	Cd in soil solution, µg L^{-1}		
					Measured	Simulated	Simulated/ Measured
I (Org)	0-25	6.0	1.6	0.11	0.10	1.09	10.9
	25-55	6.3	0.4	0.03	0.04	0.62	15.6
	55-85	6.6	0.1	0.03	0.08	1.31	16.3
II (Org)	0-25	6.2	2.7	0.11	0.15	0.57	3.8
	25-55	5.2	0.8	0.03	0.18	1.31	7.3
	55-85	5.0	0.6	0.03	1.68	2.08	1.2
III (Conv)	0-25	6.5	5.3	0.11	0.05	0.23	4.7
	25-55	4.6	1.5	0.03	0.05	1.61	32.2
	55-85	4.1	1.3	0.02		2.16	
IV (Org)	0-25	5.7	4.6	0.10	0.05	0.61	12.2
	25-55	4.6	2.2	0.03	0.10	1.19	11.9
	55-85	4.0	1.0	0.01		1.50	
Mean							*11.6*

1.8.2. Data from long term field trials in France

To evaluate Cd leaching under annual crops in France, we used data from the QualiAgro and PRO'spective long term field experiments. These form part of the SOERE-PRO (network of long-term experiments dedicated to the study of impacts of organic waste product recycling) certified by ALLENVI (Alliance Nationale de Recherche pour l'Environnement) and integrated as a department of the ''Investment d'Avenir'' infrastructure, known as AnaEE-France, overseen by the French National Research Agency (ANR-11-INBS-0001). The SOERE-PRO is under the scientific supervision of Dr Sabine Houot.

1.8.2.1. QualiAgro trial in Feucherolles

The experimental data of QualiAgro came from Cambier et al. (2014) and Filipović et al. (2016) who measured Cd water concentrations and fluxes for nearly six years, using a lysimeter sampling system at a 45 cm depth, in three plots cultivated with maize and wheat. One was a control plot (CONT), the other two receiving either sewage sludge compost (SGW) or municipal waste compost (MSW). This long-term field experiment was located at Feucherolles, 35 km west of Paris (France), on a silt loam Luvisol.

For each plot, Cambier et al. (2014) give the composition of the soil layers of 0-28 cm, 28-35 cm, 35-50 cm and 50-90 cm depth. From these data, we estimated the average composition of the 28-45 cm and 0-45 cm layers. From the properties of the 0-28 cm, 28-45 cm and 0-45 cm, we estimated the concentration in the soil solution ($[Cd]_w$) and the amount leached (Q_{lea}) according to the model proposed by Six and Smolders (2014):

$$Q_{lea} = 10\, W_{lea}\, [Cd]_w$$

$$[Cd]_w = \frac{[Cd]_s}{K_D}$$

$$\log(K_D) = -0.94 + 0.51(pH_{water} - 0.54) + 0.79\log(C_{org})$$

W_{lea} is the annual precipitation excess (corresponding here to 179 mm), pH_{water} is the soil pH in water suspension, C_{org} is the soil organic C content (% mass) and $[Cd]_s$ is the total soil Cd content.

$[Cd]_w$ estimated from each layer, was compared to the mean concentration measured in the water percolating at 45 cm in 2011-2013 and in 2012-2013. Q_{lea} was compared to the measured amount of Cd leached during 68 months, as given by Filipović et al. (2016) (Table S10).

1.8.2.2. PRO'spective trial in Colmar

The PRO'spective field experiment is conducted by INRA Colmar in collaboration with SMRA68 (Syndicat Mixte Recyclage Agricole du Haut-Rhin), ARAA (Association pour la Relance Agronomique en Alsace) and UHA (Université de Haute-Alsace). The field experiment is financed by SMRA68, ADEME, AERM, Veolia, SITEUCE, Arvalis, Terralys, SEDE, COVED, SM4. It is located in the Alsace region, near Colmar.

Cadmium concentrations and fluxes were measured for 5.5 years using two lysimeters per plot to quantify and analyse the solution percolating at a 45 cm depth, in six plots cultivated with grain maize, winter wheat, sugar beet and spring barley. One of the plots was a control plot without amendment, the other five receiving either sewage sludge, composted sewage sludge, composted biowaste, bovine manure and composted manure.

From the extensive data set provided by Denis Montenach (INRA, Colmar), we calculated the summary presented in

Table **S*11***. Cadmium concentrations in soil solutions were measured by ICP-MS, with a detection limit of 0.015 µg L^{-1}. Values below this limit were replaced by half the limit.

Table S10. Comparison of simulated Cd concentrations in percolating water and of simulated leached Cd to values measured in the QualiAgro long-term field experiment in Feucherolles (France). Measured values come from Cambier et al. (2014) and Filipović et al. (2016).

Plot	**Soil layer cm**	**Total Cd mg kg^{-1}**	**pH water**	**Organic C %**	**Cd concentration in soil solution**					**Leached Cd**		
					Measured at 45 cm μg L^{-1}		**Simulated μg L^{-1}**	**Simulated/Measured**		**Measured at 45 cm g ha^{-1} yr^{-1}**	**Simulated g ha^{-1} yr^{-1}**	**Simulated/ Measured**
					Mean 2011-2013	**Mean 2012-2013**	**Simulated**	**2011-2013**	**2012-2013**			
SGW	0-28	0.223	6.86	1.514	0.042	0.052	0.837	19.9	16.1	0.14	1.50	10.6
MSW	0-28	0.222	7.51	1.353	0.042	0.052	0.425	10.1	8.2	0.11	0.76	7.2
CONT	0-28	0.195	6.63	1.012	0.042	0.052	1.318	31.4	25.4	0.11	2.36	22.3
SGW	28-45	0.159	7.06	0.744	0.042	0.052	0.822	19.6	15.8	0.14	1.47	10.4
MSW	28-45	0.169	7.63	0.743	0.042	0.052	0.448	10.7	8.6	0.11	0.80	7.6
CONT	28-45	0.145	6.95	0.638	0.042	0.052	0.965	23.0	18.6	0.11	1.73	16.3
SGW	0-45	0.199	6.94	1.223	0.042	0.052	0.807	19.2	15.5	0.14	1.44	10.2
MSW	0-45	0.202	7.56	1.123	0.042	0.052	0.424	10.1	8.1	0.11	0.76	7.2
CONT	0-45	0.176	6.75	0.871	0.042	0.052	1.162	27.7	22.3	0.11	2.08	19.6
Mean	0-45							*19.0*	*15.3*			*12.3*

Table S11. Comparison of simulated Cd concentrations in percolating water and of simulated leached Cd to values measured in the PRO'spective long-term field experiment in Colmar (France). Cd concentrations measured in soil solution result from an averaging of 365 measurements made in 38 drainage episodes from January 2010 until July 2016. K_d, Cd concentrations in soil solutions and Cd fluxes were simulated as described in 1.8.2.1.

Amendment	**Plot number**	**Total Cd**	**Organic C**	**pH water**	**Simulated K_d**	**Cd concentration in soil solution**			**Cumulated Cd fluxes**		
						Simulated	**Measured at 45 cm**	**Simulated /Measured**	**Simulated**	**Measured at 45 cm**	**Simulated /Measured**
		mg kg^{-1}	**%**		**L kg^{-1}**	**µg L^{-1}**			**g ha^{-1} yr^{-1}**		
Sewage sludge	I T101	0.238	1.25	8.52	1608	0.148	0.039	3.8	0.071	0.016	4.4
Composted sludge	I T102	0.241	1.38	8.43	1565	0.154	0.060	2.6	0.092	0.024	3.8
Composted biowaste	I T103	0.236	1.42	8.47	1677	0.141	0.033	4.3	0.061	0.011	5.4
Manure	I T104	0.242	1.36	8.47	1621	0.149	0.040	3.8	0.143	0.026	5.6
Composted manure	I T105	0.240	1.35	8.45	1574	0.152	0.046	3.3	0.208	0.033	6.4
None (control)	I T106	0.240	1.19	8.50	1511	0.159	0.046	3.5	0.137	0.027	5.1
Mean		**0.240**	**1.33**	**8.47**	**1593**	**0.151**	**0.044**	**3.5**	**0.119**	**0.023**	**5.1**

1.9. Cadmium crop offtake

Here are presented the data and calculations used to derive the transfer factors (TF) for each crop. Data from France were used in priority, when available. When several values were available, the mean was calculated.

1.9.1. Barley

We did not find data concerning Cd content in barley grain from France. From the work of Kaniuczack et al. (2011) it could be calculated that in Poland, spring barley grains contained 1.33 times more Cd than wheat grains produced on the same soil. A similar calculation made with the values given by Wiersma et al. (1986) for the Netherlands lead to a factor of 1.86. Averaging these two factors gave a value of 1.6. Therefore, the TF of barley was obtained by multiplying that of wheat by 1.6, which gave 0.24.

1.9.2. Durum wheat

According to the data of Baize et al. (2003) obtained in field trials with durum wheat and wheat near Nîmes and Toulouse, median Cd contents in wheat grains range between 0.042 and 0.053 mg Cd (kg DM)$^{-1}$ (the mean of the medians being 0.0475 mg Cd (kg DM)$^{-1}$) and in durum wheat between 0.108 and 0.196 mg Cd (kg DM$^{)-1}$ (the mean median is 0.152 mg Cd (kg DM$^{)-1}$). The ratio of the median mean of durum wheat to that of wheat (0.152/0.0475) is 3.2. Taking a TF of 0.15 for wheat (see below), the TF for durum wheat is 0.15 x 3.2 = 0.48.

Wu et al. (2002) found an average of 0.182 mg Cd (kg DM)$^{-1}$ in durum wheat growing in a soil containing 0.34 mg Cd (kg DM)$^{-1}$. After a correction of dry matter, TF = 0.1547/0.34 = 0.455.

As we had little data for durum wheat, we averaged the previous values, which gave TF = 0.47.

1.9.3. Field pea

Engqvist and Mårtensson (2005) measured the Cd content in the seeds of 15 field pea cultivars grown on five French sites. The mean TF is 0.13, taking a dry matter content of 84% (COMIFER, 2009).

1.9.4. Forage maize

As the yield of forage maize was given on a dry matter basis, the TF for this crop was calculated on the same basis. Wiersma et al. (1986) found a mean Cd content of 0.43 mg Cd (kg DM)$^{-1}$ in forage maize produced in a soil containing 0.43 mg Cd (kg DM)$^{-1}$, which gives TF = 1.08. Smolders et al. (2007) found a median content of 0.29 in forage maize grown in soil containing 1.0, which makes a TF of 0.29. We used the mean of these two values, i.e. TF = 0.69.

1.9.5. Grain maize

Various data were found in the literature, showing the high variability of the Cd transfer factor to maize grain (***Table S12***), from which a mean value of 0.11 was calculated.

Table S12. Values of the Cd transfer factor (TF) to maize grain, derived from the literature, using a grain moisture content of 15%. Cadmium contents are in mg (kg DM)$^{-1}$.

Reference	Cd in grain	Cd in soil	TF
Baize et al. (2006)	0.01	0.24	0.035
Carbonell et al. (2011)			0.034
Guo et al. (2011)	0.263, 0.486, 0.311 according to the cultivar	0.96	0.23, 0.43, 0.28.
Lavado et al. (2007)	0.02	0.79	0.022
Li et al. (2012)			0.031, 0.037
Mench et al. (1994), figure 1	0.25	1.3	0.16
Stanislawska-Glubiak et al. (2015)			0.006, 0.028
Wang et al. (2014)	0.031	0.148	0.018
Wang et al. (2016)		1.64	0.077 to 0.17

1.9.6. Potato

Based on the analysis of 100 samples, Gravoueille et al. (2010) reported a mean Cd content in potato tubers of 0.023 mg Cd (kg FW)$^{-1}$. Divided by the mean soil Cd content (0.31 mg Cd (kg DM)$^{-1}$), this gave a TF of 0.074.

From a moderately contaminated area in Belgium and Netherlands, Smolders et al. (2007) found a median content of 0.14 mg Cd (kg DM)$^{-1}$ in tubers, for a median soil content of 0.34 mg Cd (kg DM)$^{-1}$. Considering a 22% dry matter content (Wiersma et al., 1986), these data gave a TF of 0.091.

In this study, we used a mean of these two values, i.e. a TF of 0.083 for potatoes.

1.9.7. Rape

Sylvie Dauguet from Terres Inovia provided us with the data represented in Figure 3 from in Dauguet and Lacoste (2013). The mean moisture content of rape seeds is 9% and the mean Cd content is 0.047 mg Cd (kg DM)$^{-1}$. With a mean soil content of 0.31 mg Cd (kg DM)$^{-1}$, this gives a TF of 0.14.

1.9.8. Sugar beet

In a study carried out by the Institut Technique de la Betterave (ITB), the following relationship was established:

Cdroot = 0.0549 Cdsoil +0.4964, $r^2 = 0.45$

where Cdroot and Cdsoil are the Cd contents in roots and soils, respectively (mg Cd (kg DM)$^{-1}$).

Taking a Cdsoil = 0.31 mg Cd (kg DM)$^{-1}$ and a dry matter content in the root of 25%, this relationship gave a TF = 0.41

1.9.9. Sunflower

Sylvie Dauguet from Terres Inovia provided us with the data represented in Figure 3 by Dauguet and Lacoste (2013). The mean moisture content of sunflower 9% and the mean Cd content is 0.358 mg Cd (kg DM)$^{-1}$. The mean soil content was 0.26 mg Cd (kg DM)$^{-1}$ when considering the regions were sunflower is the most cultivated (BDETM 2000-2010, 'departments' of Aquitaine, Centre, Languedoc-Roussillon, Midi, Poitou-Charentes). Calculations gave a TF of 1.25.

1.9.10. Triticale

Because of a lack of data concerning triticale, the TF of wheat was used for this crop.

1.9.11. Wheat

Denaix et al. (2010) made a review of Cd content in French wheat grain. They found a mean value of 0.054 mg Cd (kg DM)$^{-1}$. Considering that the grain contains 85% dry matter (Wiersma et al., 1986) and that the mean soil content is 0.31 mg Cd (kg DM)$^{-1}$, TF is then 0.15. This value is close to that (0.14) used by Six and Smolders (2014) for Cd balance at the European level.

1.10. Crop rotations

1.10.1. Alsace

Grain maize, grain maize, wheat, grain maize, grain maize, wheat, grain maize, grain maize, wheat, grain maize, grain maize, wheat, grain maize, grain maize, sugar beet, grain maize, forage maize.

1.10.2. Aquitaine

Grain maize, grain maize, grain maize, sunflower, grain maize, grain maize, forage maize, wheat.

1.10.3. Auvergne

Forage maize, wheat, barley, grain maize, wheat, forage maize, wheat, triticale, grain maize, wheat, triticale, sunflower, wheat, rape, wheat.

1.10.4. Bourgogne

Forage maize, wheat, barley, rape, wheat, triticale, rape, wheat, rape, wheat, barley, rape, wheat, barley, rape, wheat, grain maize, wheat, barley, wheat, barley, rape, wheat, sunflower, wheat, barley.

1.10.5. Bretagne

Forage maize, wheat, barley, forage maize, grain maize, wheat, triticale, rape, wheat, forage maize, wheat, forage maize, wheat, forage maize, forage maize, wheat, grain maize

1.10.6. Centre

Sunflower, durum wheat, wheat, grain maize, wheat, barley, rape, wheat, wheat, barley, rape, wheat, barley, grain maize, rape, wheat, wheat, rape, wheat.

1.10.7. Champagne-Ardenne

Sugar beet, wheat, barley, rape, wheat, grain maize, wheat, barley, rape, wheat, barley, wheat, sugar beet, wheat, barley, rape, wheat, rape, wheat, barley, forage maize, wheat, barley.

1.10.8. Corsica

Grain maize, grain maize, wheat, forage maize, grain maize, triticale, grain maize, grain maize, triticale, grain maize, forage maize, barley, grain maize, grain maize, barley, grain maize, barley.

1.10.9. Franche-Comté

Sunflower, wheat, barley, forage maize, wheat, rape, grain maize, wheat, barley, forage maize, wheat, rape, grain maize, wheat, barley, grain maize, wheat, rape, wheat, barley, grain maize, wheat, rape, wheat, triticale.

1.10.10. Île de France

Sugar beet, wheat, barley, rape, wheat, barley, grain maize, wheat, wheat, rape, wheat, wheat.

1.10.11. Languedoc-Roussillon

Sunflower, durum wheat, wheat, durum wheat, sunflower, durum wheat, durum wheat, sunflower, durum wheat, barley, durum wheat, sunflower, durum wheat, triticale, durum wheat, sunflower, durum wheat, durum wheat, rape, barley, durum wheat, durum wheat.

1.10.12. Limousin

Forage maize, wheat, triticale, forage maize, wheat, triticale, forage maize, wheat, triticale, forage maize, triticale, barley, forage maize, wheat, triticale, grain maize, barley.

1.10.13. Lorraine

Forage maize, wheat, barley, rape, wheat, barley, rape, wheat.

1.10.14. Midi-Pyrénées

Sunflower, wheat, grain maize, barley, sunflower, wheat, grain maize, barley, sunflower, wheat, sunflower, wheat, grain maize, durum wheat, sunflower, wheat, rape, durum wheat, forage maize, triticale.

1.10.15. Nord-Pas de Calais

Potato, wheat, barley, sugar beet, wheat, barley, potato, wheat, wheat, sugar beet, wheat, wheat, forage maize, wheat, rape, wheat, forage maize, wheat, forage maize, wheat.

1.10.16. Basse-Normandie

Forage maize, wheat, barley, forage maize, wheat, forage maize, wheat, forage maize, wheat, forage maize, wheat, rape, wheat.

1.10.17. Haute-Normandie

Sugar beet, wheat, barley, potato, wheat, barley, forage maize, wheat, wheat, forage maize, wheat, wheat, rape, wheat, forage maize, wheat, rape, wheat, wheat, rape, wheat, wheat, rape, wheat.

1.10.18. Pays de Loire

Sunflower, wheat, forage maize, barley, grain maize, wheat, forage maize, triticale, forage maize, wheat, grain maize, wheat, forage maize, wheat, forage maize, wheat, grain maize, wheat, forage maize, wheat, forage maize, rape, wheat.

1.10.19. Picardie

Sugar beet, wheat, barley, rape, wheat, wheat, sugar beet, wheat, rape, wheat, forage maize, wheat, wheat, sugar beet, wheat, barley, rape, wheat, grain maize, wheat, potato, wheat, wheat.

1.10.20. Poitou-Charentes

Grain maize, wheat, barley, sunflower, wheat, rape, grain maize, wheat, sunflower, wheat, barley, grain maize, wheat, sunflower, wheat, grain maize, durum wheat, wheat, sunflower, wheat, triticale, forage maize, wheat, rape.

1.10.21. Provence-Alpes-Côte d'Azur

Sunflower, durum wheat, durum wheat, wheat, durum wheat, grain maize, durum wheat, durum wheat, barley, durum wheat, wheat, durum wheat, sunflower, durum wheat, durum wheat, barley, durum wheat, rape, durum wheat, durum wheat, triticale, durum wheat, barley.

1.10.22. Rhône-Alpes

Forage maize, triticale, grain maize, wheat, grain maize, wheat, grain maize, wheat, forage maize, barley, grain maize, wheat, rape, grain maize, wheat, grain maize, wheat, forage maize, barley, sunflower.

1.10.23. France

Conventional agriculture: Sugar beet, wheat, durum wheat, grain maize, wheat, triticale, sunflower, wheat, barley, grain maize, wheat, barley, sunflower, wheat, barley, grain maize, wheat, barley, forage maize, wheat, rape, grain maize, wheat, rape, forage maize, wheat, rape, wheat, forage maize, wheat, rape, wheat.

Organic agriculture: Sugar beet, wheat, durum wheat, grain maize, field pea, wheat, triticale, sunflower, wheat, field pea, barley, grain maize, wheat, barley, field pea, sunflower, wheat, barley, grain maize, field pea, wheat, barley, forage maize, wheat, field pea, rape, grain maize, wheat, rape, field pea, forage maize, wheat, rape, wheat, field pea, forage maize, wheat, rape, wheat, field pea.

2. Supplementary data concerning the results and the discussion

Table S13. Evolution of the mean Cd content of the ploughed layer (25 cm) of soils under annual crops in France, according to different scenarios. This value is the mean of the contents calculated for each of the 22 French regions.

Scenario	**Soil Cd content, mg kg^{-1}**				**Variation, %**		
	Initial	**10 yrs**	**20 yrs**	**100 yrs**	**10 yrs**	**20 yrs**	**100 yrs**
CPA: Current P application rates	0.31	0.304	0.298	0.262	-1.9	-3.7	-15.8
CPA$_{L12}$: Same as CPA, with leaching rate /12	0.31	0.311	0.313	0.328	0.6	1.5	7.5
GPPA: P application according to good practices	0.31	0.303	0.297	0.258	-2.1	-4.2	-17.6
GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.31	0.310	0.312	0.323	0.4	1.0	5.2
EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.31	0.303	0.296	0.243	-2.1	-4.6	-22.4
EUR$_{L12}$: Same as EUR, with leaching rate /12	0.31	0.310	0.310	0.307	0.5	0.5	0.0
GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.31	0.303	0.295	0.241	-2.3	-5.0	-23.3
GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.31	0.310	0.310	0.304	0.2	0.2	-1.2

Table S14. Evolution of the mean Cd content of the ploughed layer (25 cm) of the French mean soil under annual crops, according to different scenarios taking into account the European regulation proposal limiting Cd content in P fertilizers (European Commisison, 2016) and its subsequent amendment by the European Parliament (European Parliament, 2017).

Scenario	**Soil Cd content, mg kg^{-1}**				**Variation, %**		
	Initial	**10 yrs**	**20 yrs**	**100 yrs**	**10 yrs**	**20 yrs**	**100 yrs**
EUR$_{L6}$: Current P application rates with EU regulation proposed in 2016	0.310	0.3085	0.3095	0.2982	-0.5	-0.2	-3.8
EUR$_{L6}$': Current P application rates with EU regulation amended in 2017	0.310	0.3088	0.3104	0.2990	-0.4	0.1	-3.6
GPEU$_{L6}$: P application according to good practices with EU regulation proposed in 2016	0.310	0.3078	0.3071	0.2940	-0.7	-0.9	-5.2
GPEU$_{L6}$': P application according to good practices with EU regulation amended in 2017	0.310	0.3079	0.3076	0.2945	-0.7	-0.8	-5.0
OAEU$_{L6}$: Organic agriculture, with EU regulation proposed in 2016	0.310	0.3082	0.3080	0.2985	-0.6	-0.6	-3.7
OAEU$_{L6}$': Organic agriculture, with EU regulation amended in 2017	0.310	0.3083	0.3085	0.2989	-0.5	-0.5	-3.6

Table S15. Evolution of the mean Cd content of the ploughed layer (25 cm) of the soils under annual crops in 22 French regions, according to different scenarios.

Region	**Scenario**	**Cd content, mg kg^{-1}**				**Variation, %**		
		Initial	**10 yrs**	**20 yrs**	**100 yrs**	**10 yrs**	**20 yrs**	**100 yrs**
Alsace	CPA: Current P application rates	0.270	0.272	0.271	0.276	0.7	0.4	2.1
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.270	0.277	0.282	0.328	2.5	4.4	21.5
	GPPA: P application according to good practices	0.270	0.270	0.267	0.255	-0.1	-1.2	-5.5
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.270	0.270	0.275	0.306	0.0	1.8	13.2
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.270	0.271	0.266	0.237	0.3	-1.5	-12.1
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.270	0.276	0.276	0.286	2.1	2.4	6.1
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.270	0.269	0.263	0.229	-0.3	-2.5	-15.4
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.270	0.274	0.274	0.276	1.5	1.3	2.4
Aquitaine	CPA: Current P application rates	0.190	0.190	0.189	0.185	-0.2	-0.4	-2.5
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.190	0.196	0.203	0.253	3.2	6.8	33.3
	GPPA: P application according to good practices	0.190	0.186	0.182	0.156	-2.0	-4.0	-18.1
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.190	0.193	0.196	0.218	1.4	3.1	14.6
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.190	0.189	0.185	0.154	-0.7	-2.7	-18.9
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.190	0.195	0.199	0.217	2.8	4.5	14.0
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.190	0.186	0.180	0.141	-2.2	-5.1	-25.7
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.190	0.192	0.194	0.201	1.2	2.0	5.7
Auvergne	CPA: Current P application rates	0.380	0.368	0.356	0.276	-3.2	-6.4	-27.4
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.380	0.381	0.383	0.398	0.3	0.9	4.7
	GPPA: P application according to good practices	0.380	0.366	0.352	0.259	-3.6	-7.4	-31.8
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.380	0.379	0.379	0.378	-0.2	-0.2	-0.6
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.380	0.367	0.353	0.258	-3.3	-7.0	-32.2
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.380	0.381	0.381	0.376	0.2	0.2	-1.0
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.380	0.366	0.351	0.250	-3.7	-7.7	-34.2
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.380	0.379	0.378	0.367	-0.2	-0.5	-3.5
Bourgogne	CPA: Current P application rates	0.330	0.329	0.328	0.322	-0.4	-0.5	-2.5
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.330	0.334	0.339	0.374	1.1	2.6	13.3
	GPPA: P application according to good practices	0.330	0.327	0.325	0.305	-0.8	-1.5	-7.6
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.330	0.332	0.335	0.356	0.7	1.6	7.8
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.330	0.328	0.325	0.296	-0.6	-1.5	-10.3
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.330	0.333	0.335	0.346	0.9	1.6	4.9
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.330	0.327	0.323	0.289	-0.9	-2.1	-12.5
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.330	0.332	0.333	0.338	0.6	1.0	2.5

Region	**Scenario**	**Cd content, mg kg^{-1}**				**Variation, %**		
		Initial	**10 yrs**	**20 yrs**	**100 yrs**	**10 yrs**	**20 yrs**	**100 yrs**
Bretagne	CPA: Current P application rates	0.210	0.204	0.196	0.154	-2.8	-6.5	-26.7
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.210	0.212	0.213	0.227	1.1	1.5	7.9
	GPPA: P application according to good practices	0.210	0.203	0.197	0.154	-3.2	-6.4	-26.6
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.210	0.211	0.213	0.227	0.7	1.6	8.0
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.210	0.204	0.195	0.142	-3.0	-7.3	-32.3
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.210	0.212	0.212	0.212	0.9	0.7	1.1
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.210	0.203	0.194	0.139	-3.4	-7.5	-33.8
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.210	0.211	0.211	0.209	0.5	0.5	-0.7
Centre	CPA: Current P application rates	0.250	0.250	0.251	0.253	0.0	0.3	1.1
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.250	0.254	0.259	0.296	1.6	3.6	18.4
	GPPA: P application according to good practices	0.250	0.250	0.252	0.258	0.2	0.7	3.3
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.250	0.254	0.260	0.302	1.8	4.1	20.9
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.250	0.249	0.247	0.227	-0.3	-1.1	-9.3
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.250	0.253	0.256	0.268	1.3	2.3	7.2
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.250	0.250	0.248	0.229	-0.1	-0.8	-8.3
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.250	0.254	0.257	0.271	1.5	2.6	8.3
Champagne-Ardenne	CPA: Current P application rates	0.450	0.448	0.445	0.428	-0.5	-1.0	-4.9
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.450	0.451	0.452	0.459	0.2	0.4	2.1
	GPPA: P application according to good practices	0.450	0.447	0.444	0.422	-0.7	-1.3	-6.3
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.450	0.450	0.450	0.453	0.0	0.1	0.6
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.450	0.447	0.441	0.397	-0.7	-1.9	-11.9
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.450	0.450	0.448	0.427	0.0	-0.5	-5.2
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.450	0.446	0.440	0.394	-0.9	-2.2	-12.5
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.450	0.449	0.447	0.424	-0.2	-0.7	-5.9
Corse	CPA: Current P application rates	0.310	0.295	0.280	0.190	-4.9	-9.6	-38.8
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.310	0.312	0.315	0.331	0.6	1.5	6.9
	GPPA: P application according to good practices	0.310	0.297	0.283	0.202	-4.3	-8.6	-34.8
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.310	0.314	0.318	0.348	1.2	2.6	12.3
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.310	0.294	0.277	0.170	-5.0	-10.6	-45.0
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.310	0.311	0.311	0.306	0.4	0.5	-1.2
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.310	0.296	0.279	0.176	-4.6	-9.9	-43.3
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.310	0.313	0.314	0.314	0.9	1.2	1.2

Region	**Scenario**	**Cd content, mg kg^{-1}**				**Variation, %**		
		Initial	**10 yrs**	**20 yrs**	**100 yrs**	**10 yrs**	**20 yrs**	**100 yrs**
Franche-Comté	CPA: Current P application rates	0.500	0.468	0.435	0.254	-6.5	-13.0	-49.1
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.500	0.500	0.501	0.509	0.1	0.3	1.7
	GPPA: P application according to good practices	0.500	0.465	0.430	0.235	-7.0	-14.1	-52.9
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.500	0.498	0.496	0.482	-0.5	-0.8	-3.6
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.500	0.467	0.432	0.234	-6.6	-13.7	-53.3
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.500	0.500	0.498	0.480	0.0	-0.4	-3.9
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.500	0.464	0.428	0.225	-7.1	-14.4	-54.9
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.500	0.497	0.494	0.468	-0.5	-1.1	-6.4
Île de France	CPA: Current P application rates	0.290	0.287	0.284	0.263	-1.1	-2.2	-9.3
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.290	0.290	0.290	0.293	-0.1	0.0	1.2
	GPPA: P application according to good practices	0.290	0.288	0.286	0.278	-0.7	-1.3	-4.2
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.290	0.291	0.293	0.309	0.4	0.9	6.6
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.290	0.286	0.282	0.249	-1.3	-2.9	-14.2
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.290	0.289	0.288	0.278	-0.2	-0.7	-4.0
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.290	0.287	0.284	0.255	-0.9	-2.2	-11.9
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.290	0.291	0.290	0.285	0.2	0.0	-1.6
Languedoc-Roussillon	CPA: Current P application rates	0.290	0.290	0.290	0.289	-0.1	-0.1	-0.3
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.290	0.293	0.297	0.324	1.0	2.3	11.8
	GPPA: P application according to good practices	0.290	0.289	0.290	0.289	-0.2	-0.2	-0.4
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.290	0.293	0.296	0.324	0.9	2.2	11.6
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.290	0.289	0.287	0.268	-0.3	-1.0	-7.6
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.290	0.292	0.294	0.302	0.8	1.4	4.1
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.290	0.289	0.287	0.268	-0.4	-1.1	-7.7
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.290	0.292	0.294	0.301	0.7	1.3	3.9
Limousin	CPA: Current P application rates	0.270	0.252	0.234	0.135	-6.7	-13.5	-50.1
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.270	0.269	0.269	0.265	-0.3	-0.5	-2.0
	GPPA: P application according to good practices	0.270	0.251	0.232	0.128	-7.0	-14.1	-52.6
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.270	0.268	0.267	0.255	-0.6	-1.1	-5.5
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.270	0.252	0.232	0.125	-6.8	-14.0	-53.6
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.270	0.269	0.267	0.252	-0.4	-1.1	-6.8
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.270	0.251	0.231	0.122	-7.1	-14.4	-54.8
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.270	0.268	0.266	0.247	-0.7	-1.5	-8.5

Region	Scenario	Cd content, mg kg^{-1}				Variation, %		
		Initial	10 yrs	20 yrs	100 yrs	10 yrs	20 yrs	100 yrs
Lorraine	CPA: Current P application rates	0.270	0.268	0.267	0.256	-0.7	-1.2	-5.1
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.270	0.273	0.276	0.304	1.0	2.4	12.7
	GPPA: P application according to good practices	0.270	0.267	0.264	0.243	-1.2	-2.3	-9.9
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.270	0.271	0.274	0.290	0.6	1.3	7.5
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.270	0.268	0.264	0.236	-0.9	-2.2	-12.8
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.270	0.272	0.274	0.282	0.8	1.4	4.5
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.270	0.266	0.262	0.230	-1.3	-2.9	-14.9
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.270	0.271	0.272	0.276	0.4	0.7	2.1
Midi-Pyrénées	CPA: Current P application rates	0.270	0.262	0.254	0.203	-2.9	-5.8	-24.8
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.270	0.273	0.276	0.302	1.1	2.4	11.7
	GPPA: P application according to good practices	0.270	0.261	0.251	0.188	-3.5	-7.2	-30.5
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.270	0.271	0.273	0.282	0.4	0.9	4.6
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.270	0.262	0.251	0.182	-3.1	-7.0	-32.7
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.270	0.272	0.273	0.276	0.9	1.2	2.2
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.270	0.260	0.249	0.175	-3.6	-7.9	-35.2
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.270	0.271	0.271	0.267	0.3	0.2	-1.0
Nord-Pas de Calais	CPA: Current P application rates	0.430	0.424	0.415	0.364	-1.4	-3.4	-15.3
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.430	0.428	0.425	0.407	-0.4	-1.2	-5.3
	GPPA: P application according to good practices	0.430	0.422	0.413	0.353	-1.8	-4.0	-17.8
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.430	0.427	0.422	0.396	-0.7	-1.8	-8.0
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.430	0.423	0.413	0.346	-1.6	-4.0	-19.5
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.430	0.428	0.422	0.388	-0.5	-1.8	-9.8
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.430	0.422	0.411	0.341	-1.9	-4.4	-20.7
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.430	0.427	0.421	0.383	-0.8	-2.2	-11.0
Basse-Normandie	CPA: Current P application rates	0.210	0.204	0.198	0.159	-2.9	-5.7	-24.1
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.210	0.211	0.212	0.222	0.5	1.1	5.7
	GPPA: P application according to good practices	0.210	0.202	0.195	0.147	-3.6	-7.2	-29.9
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.210	0.209	0.209	0.208	-0.3	-0.4	-1.2
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.210	0.204	0.196	0.145	-3.1	-6.7	-30.9
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.210	0.211	0.210	0.205	0.2	0.1	-2.3
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.210	0.202	0.194	0.140	-3.7	-7.7	-33.5
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.210	0.209	0.208	0.199	-0.4	-0.9	-5.4

Region	**Scenario**	**Cd content, mg kg^{-1}**				**Variation, %**		
		Initial	**10 yrs**	**20 yrs**	**100 yrs**	**10 yrs**	**20 yrs**	**100 yrs**
Haute-Normandie	CPA: Current P application rates	0.330	0.320	0.312	0.255	-3.0	-5.4	-22.7
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.330	0.329	0.330	0.333	-0.5	0.0	0.8
	GPPA: P application according to good practices	0.330	0.320	0.313	0.260	-3.0	-5.1	-21.1
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.330	0.328	0.331	0.338	-0.5	0.2	2.6
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.330	0.320	0.310	0.240	-3.2	-6.0	-27.4
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.330	0.328	0.328	0.315	-0.6	-0.7	-4.5
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.330	0.320	0.311	0.242	-3.2	-5.9	-26.8
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.330	0.328	0.328	0.317	-0.6	-0.6	-3.8
Pays de Loire	CPA: Current P application rates	0.220	0.214	0.207	0.165	-2.9	-5.8	-24.9
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.220	0.220	0.221	0.227	0.2	0.6	3.1
	GPPA: P application according to good practices	0.220	0.213	0.206	0.157	-3.3	-6.6	-28.5
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.220	0.220	0.219	0.217	-0.2	-0.3	-1.2
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.220	0.213	0.206	0.156	-3.0	-6.4	-28.9
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.220	0.220	0.220	0.216	0.1	0.0	-1.6
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.220	0.213	0.205	0.153	-3.3	-6.9	-30.6
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.220	0.219	0.219	0.212	-0.3	-0.6	-3.5
Picardie	CPA: Current P application rates	0.380	0.372	0.366	0.320	-2.2	-3.6	-15.8
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.380	0.375	0.372	0.347	-1.4	-2.1	-8.8
	GPPA: P application according to good practices	0.380	0.375	0.372	0.350	-1.3	-2.0	-8.0
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.380	0.378	0.378	0.377	-0.6	-0.4	-0.8
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.380	0.371	0.365	0.308	-2.2	-4.1	-19.0
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.380	0.374	0.370	0.334	-1.5	-2.5	-12.2
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.380	0.374	0.369	0.321	-1.6	-3.0	-15.6
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.380	0.377	0.375	0.347	-0.8	-1.4	-8.7
Poitou-Charentes	CPA: Current P application rates	0.500	0.495	0.490	0.449	-0.9	-2.0	-10.1
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.500	0.502	0.503	0.514	0.4	0.6	2.7
	GPPA: P application according to good practices	0.500	0.496	0.490	0.452	-0.9	-2.0	-9.6
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.500	0.502	0.503	0.516	0.4	0.7	3.2
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.500	0.495	0.488	0.433	-1.0	-2.4	-13.3
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.500	0.501	0.501	0.497	0.3	0.2	-0.7
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.500	0.495	0.488	0.434	-1.0	-2.4	-13.1
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.500	0.501	0.501	0.498	0.3	0.3	-0.5

Region	Scenario	Cd content, mg kg^{-1}				Variation, %		
		Initial	**10 yrs**	**20 yrs**	**100 yrs**	**10 yrs**	**20 yrs**	**100 yrs**
Provence-Alpes-Côte d'Azur	CPA: Current P application rates	0.270	0.270	0.270	0.272	0.1	0.2	0.7
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.270	0.273	0.275	0.298	1.0	2.0	10.2
	GPPA: P application according to good practices	0.270	0.271	0.273	0.284	0.5	1.0	5.0
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.270	0.274	0.278	0.310	1.4	2.9	14.7
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.270	0.270	0.269	0.258	-0.1	-0.5	-4.4
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.270	0.272	0.274	0.283	0.8	1.4	4.8
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.270	0.271	0.270	0.263	0.3	0.1	-2.5
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.270	0.273	0.275	0.288	1.2	2.0	6.8
Rhône-Alpes	CPA: Current P application rates	0.300	0.283	0.266	0.171	-5.7	-11.2	-43.1
	CPA$_{L12}$: Same as CPA, with leaching rate /12	0.300	0.301	0.304	0.319	0.5	1.3	6.3
	GPPA: P application according to good practices	0.300	0.282	0.264	0.161	-6.0	-12.1	-46.4
	GPPA$_{L12}$: Same as GPPA, with leaching rate /12	0.300	0.300	0.301	0.306	0.1	0.4	1.9
	EUR: Current P application rates with EU regulation limiting Cd in fertilizers	0.300	0.282	0.264	0.154	-5.8	-12.1	-48.8
	EUR$_{L12}$: Same as EUR, with leaching rate /12	0.300	0.301	0.301	0.296	0.3	0.4	-1.3
	GPEU: P application according to good practices with EU regulation limiting Cd in fertilizers	0.300	0.281	0.262	0.149	-6.2	-12.7	-50.3
	GPEU$_{L12}$: Same as GPEU, with leaching rate /12	0.300	0.300	0.299	0.290	0.0	-0.2	-3.4

Figure S1. Influential factors ranked by ANOVA F values to explain variation in soil Cd content after one century (V_{Cd}). pH: Soil pH; Wlea: precipitation excess; Corg: soil organic carbon; Mslu: mass of slurry; Cdst0: initial soil Cd content; Y: crop yield; TF: transfer factor; Mman: mass of manure; Mafi: mass of sludge and effluents from agri-food industry; Mcom: mass of urban compost; Mnafi: mass of sludge and effluents from other industries; Miow: mass of industrial organic wastes; Msew: mass of sewage sludge; RN: crop nitrogen requirement.

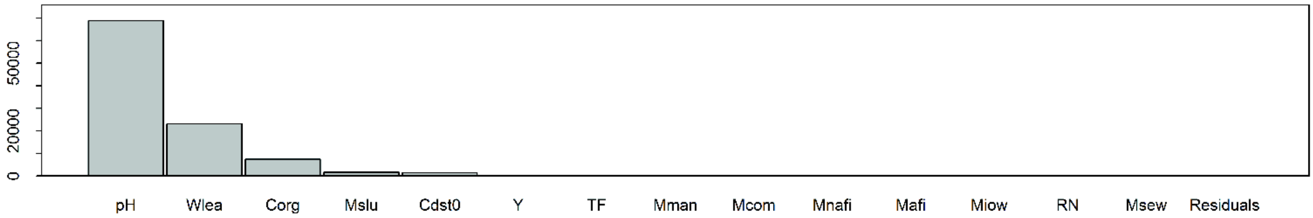

Table S16. Estimated mean Cd fluxes (in % of total inputs or outputs) in the ploughed layer (25 cm) of the mean French soil under annual crops for different scenarios.

Scenario	Inputs										Outputs	
	P fertilizers	Liming	Manure	Slurry	Urban compost	Sewage sludge	Industrial organic wastes	Sludge and effluents from agri-food-industry	Sludge and effluents from non agri-food-industry	Atmo sphere	Leaching	Crop offtake
CPA: Current P application rates	73.8	1.1	3.3	3.6	1.6	1.9	0.5	2.3	0.9	11.1	83.4	16.6
CPA$_{L6}$: Idem CPA, with leaching rate /6											53.8	46.2
CPA$_{L12}$: Idem CPA, with leaching rate /12											38.9	61.1
GPPA: P application according to good practice	65.6	1.5	4.3	4.7	2.0	2.5	0.7	3.0	1.2	14.6	83.4	16.6
GPPA$_{L6}$: Idem GPPA, with leaching rate /6											53.8	46.2
GPPA$_{L12}$: Idem GPPA, with leaching rate /12											38.9	61.1
EUR: Current P application rates with EU regulation limiting Cd in fertilizers	55.0	1.9	5.6	6.2	2.7	3.2	0.9	3.9	1.5	19.1	83.4	16.6
EUR$_{L6}$: Idem EUR, with leaching rate /6											53.8	46.2
EUR$_{L12}$: Idem EUR, with leaching rate /12											38.9	61.1
GPEU: P application according to good practice with EU regulation limiting Cd in fertilizers	46.7	2.3	6.6	7.3	3.2	3.8	1.1	4.7	1.8	22.6	83.4	16.6
GPEU$_{L6}$: Idem GPEU, with leaching rate /6											53.8	46.2
GPEU$_{L12}$: Idem GPEU, with leaching rate /12											38.9	61.1
OA: Organic agriculture	71.7	1.9	3.8	4.2	0.0	0.0	0.0	0.0	0.0	18.5	88.7	11.3
OA$_{L6}$: Same as OA, with leaching rate /6											64.4	35.6
OA$_{L12}$: Same as OA, with leaching rate /12											50.0	50.0
OAEU: Same as OA, with EU regulation limiting Cd in fertilizers	54.8	3.0	5.9	6.5	0.0	0.0	0.0	0.0	0.0	29.9	88.7	11.3
OAEU$_{L6}$: Same as OAEU, with leaching rate /6											64.4	35.6
OAEU$_{L12}$: Same as OAEU, with leaching rate /12											50.0	50.0

References

AGRESTE. Des équilibres régionaux fragiles pour l'approvisionnement en paille des litières animales. Agreste Synthèses – Animaux de boucherie. Ministère de l'Agriculture, de l'Alimentation, de la Pêche de la Ruralité et de l'Aménagement du Territoire, Montreuil-sous-Bois, 2011, pp. 4.

AGRESTE. Agreste les Dossiers n° 21 - Enquête Pratiques culturales 2011. Ministère de l'Agriculture, de l'Agroalimentaire et de la Forêt, Paris, 2014.

ARVALIS. Teneurs-seuils PK par type de sol. Compléments au recueil de fiches "Les doses et les teneurs-seuils par région" (1995). ARVALIS - Institut du Végétal, 2009, pp. 8.

ARVALIS. Fertilisation organique. ARVALIS - Institut du Végétal, Paris, 2011, https://www.arvalis-infos.fr/fertilisation-organique-@/view-8547-arvarticle.html.

ARVALIS. Plan de fumure. Intégrer les valeurs fertilisantes des produits organiques. ARVALIS - Institut du Végétal, Paris, 2016, https://www.arvalis-infos.fr/view-12357-arvarticle.html.

Baize D, Courbe C, Suc O, Schwartz C, Tercé M, Bispo A, et al. Epandages de boues d'épuration urbaines sur des terres agricoles : impacts sur la composition en éléments en traces des sols et des grains de blé tendre. Le Courrier de l'Environnement de l'INRA 2006: 35-61.

Baize D, Mench M, Sappin-Didier V, Mocquot B, Gomez A, Proix N, et al. Phytodisponibilité des éléments traces métalliques dans les grains de blé. Dossier de l'environnement de l'INRA. INRA, Paris, 2003, pp. 45-62.

Bech J, Suarez M, Reverter F, Tume P, Sánchez P, Roca N, et al. Selenium and other trace element in phosphorites: A comparison between those of the Bayovar-Sechura and other provenances. Journal of Geochemical Exploration 2010; 107: 146-160.

Bengtsson H, Alvenäs G, Nilsson SI, Hultman B, Öborn I. Cadmium, copper and zinc leaching and surface run-off losses at the Öjebyn farm in Northern Sweden—Temporal and spatial variation. Agriculture, Ecosystems & Environment 2006; 113: 120-138.

Cambier P, Pot V, Mercier V, Michaud A, Benoit P, Revallier A, et al. Impact of long-term organic residue recycling in agriculture on soil solution composition and trace metal leaching in soils. Science of The Total Environment 2014; 499: 560-573.

Carbonell G, Imperial RMd, Torrijos M, Delgado M, Rodriguez JA. Effects of municipal solid waste compost and mineral fertilizer amendments on soil properties and heavy metals distribution in maize plants (*Zea mays* L.). Chemosphere 2011; 85: 1614-1623.

COMIFER. Aide au diagnostic et à la prescription de la fertilisation phosphatée et potassique des grandes cultures. Paris: COMIFER, 1995.

COMIFER. Fertilisation PK. Grille de calcul de dose: Paris, 2009.

COMIFER. Calcul de la fertilisation azotée. Guide méthodologique pour l'établissement des prescriptions locales. Cultures annuelles et prairies. Paris La Défense: COMIFER, 2011.

COMIFER. Objectif, domaine de validité et procédure d'utilisation de la calculette permettant de calculer Mh. Notice d'utilisation (version 2), 2016.

Commission Européenne. Règlement (CE) n°889/2008 de la Commission du 5 septembre 2008 portant modalités d'application du règlement (CE) n° 834/2007 du Conseil relatif à la production biologique et à l'étiquetage des produits biologiques en ce qui concerne la production biologique, l'étiquetage et les contrôles. In: Union Européenne, editor. 889/2008, Brussels, 2008, pp. 131.

Dauguet S, Lacoste F. Les enseignements de 7 années de plan de surveillance des oléagineux. OCL. 20, 2013, pp. 119-123.

Denaix L, Nguyen C, Sappin-Didier V, Schneider A. Synthèse sur la contamination en cadmium des productions végétales de grandes cultures. Institut National de la Recherche Agronomique, RMT QUASAPROVE, 2010, pp. 11.

Engqvist G, Mårtensson A. Cadmium uptake in field pea cultivars grown under French and Swedish conditions. Acta Agriculturae Scandinavica, Section B — Soil & Plant Science 2005; 55: 64-67.

European Commisison. Proposal for a regulation of the European Parliament and of the Council laying down rules on the making available on the market of CE marked fertilising products and amending Regulations (EC) No 1069/2009 and (EC) No 1107/2009. In: European Commission, editor. COM(2016) 157 final. European Commission, Brussels, 2016, pp. 43.

European Parliament. CE marked fertilising products ***I. Amendments adopted by the European Parliament on 24 October 2017 on the proposal for a regulation of the European Parliament and of the Council laying down rules on the making available on the market of CE marked fertilising products and amending Regulations (EC) No 1069/2009 and (EC) No 1107/2009 (COM(2016)0157 – C8-0123/2016 – 2016/0084(COD)). In: European Parliament, editor. P8_TA-PROV(2017)0392. . European Parliament, Strasbourg, 2017, pp. 116.

Filipović V, Cambier P, Filipović L, Coquet Y, Pot V, Bodineau G, et al. Modeling copper and cadmium mobility in an Albeluvisol amended with urban waste composts. Vadose Zone Journal 2016; 15: 1-15.

Gilbert N. Environment: The disappearing nutrient. Nature. 461, 2009, pp. 716-718.

Gravoueille J-M, Ponthonne S, Fourrier F. Surveillance phytosanitaire : Très bonne maîtrise des teneurs résiduelles. CNIPT, 2010, pp. 3.

Guo X-F, Wei Z-B, Wu Q-T, Qiu J-R, Zhou J-L. Cadmium and zinc accumulation in maize grain as affected by cultivars and chemical fixation amendments. Pedosphere 2011; 21: 650-656.

Houot S, Pons M-N, Pradel M, Tibi A, Aubry C, Augusto L, et al. Valorisation des matières fertilisantes d'origine résiduaire sur les sols à usage agricole ou forestier. Impacts agronomiques, environnementaux, socio-économiques. Expertise scientifique collective, Paris, 2014, pp. 930.

Kaniuczack J, Hajduk E, Wlasniewski S. Effect of liming and mineral fertilization on cadmium content in grain of spring barley (*Hordeum vulgare* L.) and winter wheat (*Triticum aestivum* L.) cultivated on loessial soil. Journal of Elementology 2011: 535-542.

Lavado RS, Rodríguez M, Alvarez R, Taboada MA, Zubillaga MS. Transfer of potentially toxic elements from biosolid-treated soils to maize and wheat crops. Agriculture, Ecosystems & Environment 2007; 118: 312-318.

Li Q, Guo X-Y, Xu X-H, Zuo Y-B, Wei D-P, Ma Y-B. Phytoavailability of copper, zinc and cadmium in sewage sludge-amended calcareous soils. Pedosphere 2012; 22: 254-262.

Loyon L. Inventaire des quantités de fumier bovin stockable au champ en France. 22ème Journée Rencontres Recherches Ruminants. Institut de l'Elevage, Paris, 2015, pp. 73.

McLaughlin MJ, Tiller KG, Naidu R, Stevens DP. Review: the behaviour and environmental impact of contaminants in fertilizers. Australian Journal of Soil Research 1996; 34: 1-54.

Mench MJ, Martin E, Solda P. After effects of metals derived from a highly metal-polluted sludge on maize (*Zea mays* L.). Water, Air, and Soil Pollution 1994; 75: 277-291.

Six L, Smolders E. Future trends in soil cadmium concentration under current cadmium fluxes to European agricultural soils. Science of The Total Environment 2014; 485–486: 319-328.

Smolders E, Jansson G, Van Laer L, Ruttens A, Vangronsveld J, Römkens P, et al. Teeltadvies voor de landbouw in kader van het Interreg project BeNeKempen. Alterra, Universiteit Hasselt, Katholieke Universiteit Leuven, Bodemkundige Dienst België, CODA, 2007, pp. 39.

Stanislawska-Glubiak E, Korzeniowska J, Kocon A. Effect of peat on the accumulation and translocation of heavy metals by maize grown in contaminated soils. Environmental Science and Pollution Research 2015; 22: 4706-4714.

Wang A, Wang M, Liao Q, He X. Characterization of Cd translocation and accumulation in 19 maize cultivars grown on Cd-contaminated soil: implication of maize cultivar selection for minimal risk to human health and for phytoremediation. Environmental Science and Pollution Research 2016; 23: 5410-5419.

Wang Q, Zhang J, Zhao B, Xin X, Zhang C, Zhang H. The influence of long-term fertilization on cadmium (Cd) accumulation in soil and its uptake by crops. Environmental Science and Pollution Research 2014; 21: 10377-10385.

Wiersma D, van Goor BJ, van der Veen NG. Cadmium, lead, mercury, and arsenic concentrations in crops and corresponding soils in The Netherlands. Journal of Agricultural and Food Chemistry 1986; 34: 1067-1074.

Wu J, Norvell WA, Hopkins DG, Welch RM. Spatial variability of grain cadmium and soil characteristics in a durum wheat field. Soil Science Society of America Journal 2002; 66: 268-275.