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# Risk assessment of cadmium in mineral fertilisers - fate and effects in the food chain and the environment in Norway

Scientific opinion of the Panel on Animal Feed of the Norwegian Scientific Committee for Food and Environment

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Norwegian Scientific Committee for Food and Environment (VKM)

Po 4404 Nydalen N – 0403 Oslo Norway

Phone: +47 21 62 28 00 Email: vkm@vkm.no

vkm.no

vkm.no/english

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# Risk assessment of cadmium in mineral fertilisers — fate and effects in the food chain and the environment

### Preparation of the opinion

The Norwegian Scientific Committee for Food and Environment (Vitenskapskomiteen for mat og miljø, VKM) appointed a project group to answer the request from The Norwegian Food Safety Authority. The project group consisted of eleven persons, and a project leader from the VKM secretariat. Two external referees reviewed and commented the manuscript. The VKM Panel on Animal Feed evaluated and approved the final opinion drafted by the project group.

### **Authors of the opinion**

Members of the project group that contributed to the drafting of the opinion (in alphabetical order after chair of the project group):

Trine Eggen – Chair of the project group and member of the Panel on Animal Feed in VKM. Affiliation: 1) VKM; 2) Norwegian Institute of Bioeconomy Research (NIBIO)

Heidi Amlund – Member of the project group and member of the Panel on Contaminants in VKM. Affiliation: 1) VKM; 2) National Food Institute, Technical University of Denmark

Aksel Bernhoft - Member of the project group. Affiliation: Norwegian Veterinary Institute

Ole Martin Eklo - Member of the project group and member of the Panel on Microbial Ecology in VKM. Affiliation: 1) VKM; 2) Norwegian Institute of Bioeconomy Research (NIBIO)

Gunnar Sundstøl Eriksen - Member of the project group and member of the Panel on Contaminants in VKM. Affiliation: 1) VKM; 2) Norwegian Veterinary Institute

Belinda Flem - Member of the project group. Affiliation: The Geological Survey of Norway (NGU)

Torsten Källqvist - Member of the project group. Affiliation: Former researcher in Norwegian Institute for Water Research (NIVA)

Bal Ram Singh – Member of the project group. Affiliation: Norwegian University of Life Sciences

Eiliv Steinnes - Member of the project group. Affiliation: Norwegian University of Science and Technology

Stefan Trapp - Member of the project group. Affiliation: Department of Environmental Engineering

Anne Falk Øgaard - Member of the project group. Affiliation: Norwegian Institute of Bioeconomy Research (NIBIO)

Tron Gifstad – Project leader in the VKM secretariat. Affiliation: VKM

Members of the VKM Panel on Animal Feed that assessed and approved the manuscript: In addition to Trine Eggen these were (in alphabetical order after chair of the Panel):

Åshild Krogdahl - Chair of the Panel on Animal Feed in VKM. Affiliation: 1) VKM; 2) Norwegian University of Life Sciences

Christiane Fæste. Affiliation: 1) VKM 2) Norwegian Veterinary Institute

Erik-Jan Lock. Affiliation: 1) VKM 2) Institute of Marine Research

Live Nesse. Affiliation: 1) VKM 2) Norwegian Veterinary Institute

Einar Ringø. Affiliation: 1) VKM 2) The Artic University of Norway (UiT)

Håvard Steinshamn. Affiliation: 1) VKM 2) Norwegian Institute of Bioeconomy Research (NIBIO)

Robin Ørnsrud. Affiliation: 1) VKM 2) Institute of Marine Research

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### **Competence of VKM experts**

Persons working for VKM, either as appointed members of the Committee or as external experts, do this by virtue of their scientific expertise, not as representatives for their employers or third-party interests. The Civil Services Act instructions on legal competence apply for all work prepared by VKM.

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## Summary

**Background:** Cadmium (Cd) is a toxic trace metal that can be a risk for human and animal health and the environment. Cd occurs naturally in phosphate-rich rocks used for mineral fertiliser production.

The reason for this risk assessment was the EU revision of the Fertiliser Regulation 2003/2003, which proposed that the regulations should include limit values for potentially toxic elements for both mineral and organic fertilisers, and also for soil amendments and growth media. The Cd limit was proposed to change from 137.4 mg Cd kg $^{-1}$  P to 91.6 mg Cd kg $^{-1}$  P after 3 years and 45.8 mg Cd kg $^{-1}$  P after 12 years. This corresponds to 60, 40, and 20 mg Cd kg $^{-1}$  P $_2$ O $_5$ , respectively.

The Norwegian Food Safety Authority has asked the Norwegian Scientific Committee for Food and Environment, Panel for Animal Feed for their opinion on several aspects related to Cd in mineral phosphorous fertilisers. Among other things, they have asked for background levels of Cd in agricultural soil in Norway; the fate (mobility) of Cd in soil; exposure and effects on agricultural plants for food and feed, terrestrial and aquatic organisms, farm animals for food production and humans. Further they asked how application of mineral phosphorus (P) fertilisers under different crop rotations, affects the levels of cadmium in agricultural soil in Norway from a 1, 10, and 100-year perspective, and the risk of negative effects due to this.

**Methods:** This risk assessment has been based on previous used methodology (e.g. VKM 2009; VKM, 2014, Six and Smolders, 2014). Cd mass balance in arable soil have been predicted by comparing input of Cd via different sources related to loss of Cd via leaching and harvesting of plant biomass. The mass balance indicates if the concentration of Cd in soil declines or increases over time. Based on the current Cd concentration in soil, amount of Cd input, and fate of Cd in the environment, the concentration in the major types of crop plants, in surface water, and the change of Cd in soil over time, has been estimated. The concentration of Cd in plants used for feed and food was predicted with use of transfer factors. Exposure concentration in surface water was calculated based on predicted Cd concentrations in leaching. Four different major agricultural regions in Norway (represented by the municipalities Ås, Stange, Time, and Melhus) has been part of the assessment. For each of the areas, typical crop rotation, recommended application of fertilisers, and statistics of used liming products, is the basic of the evaluation. For application of sewage sludge, both maximum allowed amounts according to today's regulation (a worst-case scenario), and average amount based on statistics, have been included in assessment scenarios. Regionspecific parameters, such as soil characteristics, annual precipitation, and infiltration rate, are factors influencing the fate and describing the environmental transfer of Cd within the environment. In this assessment, present agricultural practise based on the most common used mineral fertilisers and measured Cd concentrations (here in the range of 25-50 mg Cd kg<sup>-1</sup> P) have been compared with application of mineral fertilisers with the three maximum

limit values (MLs) 137.4, 91.6, or 45.8 mg Cd kg-1 P mineral fertilisers. Today's regulation is 100 mg Cd kg-1 P.

**Background concentration in arable soil, environmental fate and predicted environmental concentrations (PECs):** In general, the data available for Norwegian agricultural soil indicate low natural geogenic Cd concentrations (overall median < 0.2 mg kg<sup>-1</sup> dry weight, DW). However, areas with alum shales (e.g., some areas in Stange in Hedmark county) show clearly higher values, with a maximum observed Cd concentration in agricultural soil of 3.8 mg Cd kg<sup>-1</sup> DW and an average of 1.7 mg Cd kg<sup>-1</sup> DW. Hedmark, in general, does not have higher concentrations of Cd than the other regions. The average (mean) Cd concentrations in the soil in southeastern Norway, represented by Ås, (0.28 mg kg<sup>-1</sup> DW), were higher than soil in southwestern Norway, represented by Time, (0.22 mg kg<sup>-1</sup> DW), and in Mid Norway, represented by Melhus, (0.11 mg kg<sup>-1</sup> DW).

The predicted input of Cd via mineral P fertiliser to arable soil was lowest for grass production at Time and Melhus (about 0.14-0.17 g Cd ha<sup>-1</sup> yr<sup>-1</sup>), and highest for the crop rotation of potatoes, carrots, and cereals at alum shale areas at Stange (0.99 g Cd ha<sup>-1</sup> yr<sup>-1</sup>). For cereal production at Melhus, Ås, and Stange (alum shale areas) the Cd contribution was around 0.32-0.42 g Cd ha<sup>-1</sup> yr<sup>-1</sup>.

The total removal of Cd from soil via leaching and plant harvest, leaching much higher than plant removal, has been estimated to be around 3.5 g Cd ha<sup>-1</sup> yr<sup>-1</sup> at Ås and Melhus, but as high as 18-20 g Cd ha<sup>-1</sup> yr<sup>-1</sup> at Time and in the alum shale areas at Stange. The exceptionally high predicted Cd loss in Stange is due to the high background concentration, 1.7 mg Cd kg<sup>-1</sup> DW. The very high predicted loss of Cd at Time is related to a very high leaching rate, but also due to the low sorption capacity of Cd of the soil compared with in the other regions evaluated. It is general accept that sorption of Cd probably has previous been overestimated, and adjustment of sorption properties, is a part of the explanation of higher removal from the soil, and a reduction of Cd concentration in soil over time compared to previous risk assessments.

At three of four locations, a clear decline over time has been predicted. However, it might lead to an accumulation of Cd in arable soil in locations where input Cd application rate is higher than removal rate from soil, as indicated at Ås if ML level 91.6 mg and 137.4 Cd  $\rm kg^{-1}$  P in mineral P fertiliser and maximum allowed application of sewage sludge according to the present regulation is used (a realistic worst-case scenario). It is predicted a 4 and 11% increase, respectively, in 100-year perspective. This indicates that at Ås, use of mineral fertilisers with Cd concentration up to today's Norwegian regulation (100 mg Cd  $\rm kg^{-1}$  P) together with maximum allowed sewage sludge with an average Cd concentration (0.6 mg Cd  $\rm kg^{-1}$  DW, might cause a slow increase and not decrease over time.

At alum shale areas at Stange, it is predicted Cd concentrations exceeding the ML of 100  $\mu$ g Cd kg<sup>-1</sup> fresh weigh (FW) in food crops. These exceptional high concentrations are not due to ML of Cd in mineral P fertiliser, but the background concentration of Cd in alum shale soil.

Based on the present background concentrations of Cd in soil, the PEC<sub>SW</sub> was estimated at  $0.02~\mu g$  Cd L<sup>-1</sup> at Melhus,  $0.06~\mu g$  Cd L<sup>-1</sup> at Ås and 0.08~g Cd L<sup>-1</sup> at Time. In the alum shale area at Stange, PEC<sub>SW</sub> is  $0.24~\mu g$  Cd L<sup>-1</sup>. Except for Ås, with high levels of ML in in mineral P fertilisers, the PEC<sub>SW</sub> are expected to decline with time.

### **Risk characterisation; Environment:**

<u>Environmental organisms</u>: The risk characterisation of terrestrial plants and animals, and aquatic organisms has been performed on the basis of predicted environmental concentrations (PEC) of Cd in the relevant compartments, and the predicted no-effect concentrations (PNEC) for each category of organisms. The so-called Risk Characterisation Ratio (RCR), defined as PEC/PNEC, is a quantitative expression of risk. At RCRs higher than 1, unacceptable effects on organisms are likely to occur. The higher the ratio, the more likely it is that unacceptable effects may occur. The risk characterisation, based on the present levels of Cd in soil at Stange and Ås, gave the following results:

<u>Terrestrial organisms:</u> The RCRs are below 1 for all scenarios, indicating no risk from direct exposure to soil for terrestrial organisms. The highest RCRs (0.74) are found in Stange, where the present background Cd concentration is 26% below the PNEC<sub>soil</sub>.

In most scenarios, the PECs, and consequently also the RCRs, will decline over 100 years. The effects of different Cd contents in mineral P fertilisers is initially insignificant, but increases with time. The biggest increase is found in Melhus, where the PEC and RCR after 100 years using fertilisers with the highest content of Cd in mineral P are 68% higher than using fertilisers with the present content of Cd in mineral P for 100 years. However, the RCR is still well below 1, indicating no risk of effects on terrestrial organisms.

The RCRs indicate a risk of secondary poisoning of terrestrial mammals and birds at the present level of soil Cd content at Stange. The predicted concentrations show a decline with time, but the RCRs are still above 1 after 100 years, independent of the level of Cd in fertilisers. For the Ås, Time, and Melhus scenarios, all RCRs are well below 1, indicating no risk of secondary poisoning.

<u>Agricultural plants:</u> RCR are below 1 for all scenarios, indicating no risk from direct exposure to soil for terrestrial plants even at the highest Cd content of Cd in fertilisers (137.4 mg Cd kg<sup>-1</sup> P). The effect of different Cd content in fertilisers is the same as described for terrestrial organisms.

Aquatic organisms: The estimated concentrations of Cd in surface water, indicate that the present background concentration of Cd in agricultural soils may constitute a risk of effects on aquatic organisms found in recipients of drainage water from arable land. This is particularly the case in Stange, where the RCR based on present background concentrations is 3. The RCRs decrease with time at all levels of Cd in fertiliser P, but are still above 1 after 100 years. In Time, the RCR based on present background is 0.97, but decreases rapidly with time. In Ås, the present RCR is 0.69 and increases with time to 0.75 after 100 years in

the worst-case scenario, due to accumulation of Cd in soil at this site. Melhus shows the lowest RCR (0.21).

The contribution of Cd from fertilisers has a small effect on the calculated RCRs initially, but after 100 years, an increase in Cd levels in fertilisers (from the present level to 137.4 mg Cd kg<sup>-1</sup> P), may cause an increase in RCR of as much as 68% in Melhus, but only 5-6% in Stange, where the background concentration in soil is high.

<u>Sediment-dwelling (benthic) organisms:</u> The RCR is 1, indicating a risk to sediment-dwelling organisms at Stange with the background concentration of Cd in soil, and after one year with fertiliser application. As Cd concentrations decrease with time, the RCRs are below 1 after 10 and 100 years, indicating no further risk of effects.

For all scenarios, the trends are the same as described for surface-water organisms; this is expected, as the concentrations of Cd in sediments are proportional to those in surface water.

### **Risk characterisation; Farm animals and humans:**

The estimated Cd exposure of farm animals is considered far lower than those at which toxicological data indicate adverse effects will occur, and should not be of any health concern. However, the safe exposure levels for different animal species are uncertain. Cadmium accumulates in liver and kidneys, and these organs from older animals can enter the food-chain, and Cd potentially transferred to consumers. However, there is lack of data on the quantitative relation between Cd exposure and Cd residues in these animal products.

Except for worst-case at Ås, a decline of Cd in soil and crops is predicted over time. Thus, in a long-term perspective, Cd concentrations in crop plants at Stange, Time and Melhus will decrease. At Ås, with use of maximum allowed sewage sludge and ML of 91.6 and 137.4 mg Cd kg<sup>-1</sup> P, a marginal increase over 100-years perspective, 2-3% and 11-12% respectively, is predicted.

Thus, despite use of mineral fertiliser, Cd concentrations in animal diets will decrease and reduce the risk for Cd concentrations of concern in animal products for human consumption, as well as the low risk for adverse health effects in animals, in most regions. However, prevention of liver and kidneys of older animals from entering the human food chain remains an important measure to decrease the risk of Cd from animal origin.

The current mean dietary exposure to Cd is estimated to exceed the tolerable weekly intake (TWI) for the youngest age groups and for most age groups for high consumers. A reduction in Cd concentrations in the main dietary sources, such as cereals, potatoes, and root vegetables, is therefore desirable as it will decrease the exposure and hence reduce the risk for the population.

VKM has evaluated risk for children eating soil, and, given that this exposure route usually only occurs for a limited age span and the estimated exposure is low compared to intake from the diet, considers this exposure route to be of low risk.

Cd levels in farm animals drinking water is usually far below that in their feed and in pasture plants, and is not expected to contribute significantly to their total exposure.

**<u>Key words:</u>** VKM, risk assessment, Norwegian Scientific Committee for Food and Environment, Norwegian Environment Agency, Cadmium, Mineral fertiliser.

# Sammendrag på norsk

**Bakgrunn:** Kadmium (Cd) er et giftig spormetall som kan utgjøre en risiko for miljø og helse til dyr og mennesker. Kadmium forekommer naturlig i fosfatrike bergarter som brukes til produksjon av mineralgjødsel.

Bakgrunnen for risikovurderingen var EUs revisjon av gjødselforordningen 2003/2003 som foreslo at forordningen bør inneholde grenseverdier for potensielt giftige elementer for både mineralgjødsel og gjødsel med organisk opphav, samt inkludere jordforbedringsmidler og vekstmedier. Kadmiumgrensen ble foreslått endret fra 137,4 mg Cd kg $^{-1}$  P (fosfor) til 91,6 mg Cd kg $^{-1}$  P etter 3 år, og til 45,8 mg Cd kg $^{-1}$  P etter 12 år. Dette tilsvarer henholdsvis 60, 40, og 20 mg Cd kg $^{-1}$  P $_2$ O $_5$ .

Mattilsynet har bedt Vitenskapskomiteen for mat og miljø (VKM), faggruppe for fôr, om deres vurdering av flere aspekter knyttet til kadmium i mineralsk fosforgjødsel. VKM ble blant annet bedt om å beskrive bakgrunnsnivåer av kadmium i jordbruksjord i Norge: skjebnen (mobilitet) av kadmium i jord og i miljøet, eksponering og effekter på landbruksvekster brukt til mat og fôr, terrestriske og vannlevende organismer, husdyr for matproduksjon og mennesker. Videre ble det etterspurt hvordan bruken av mineralsk fosfatgjødsel under ulike vekstrotasjoner påvirker nivåene av kadmium i jordbruksjord i Norge etter 1, 10 og 100 år, og risikoen for negative effekter på grunn av dette.

**Metoder:** Denne risikovurderingen har vært gjennomført i henhold til tidligere metodikk brukt for tilsvarende risikovurderinger (for eksempel VKM 2009, VKM 2014, Six and Smolders 2014). Massebalanse i jord har blitt estimert ved å sammenligne tilført kadmium, og tap av kadmium via utlekking og høsting av avlinger. En slik massebalanse indikerer om konsentrasjonen av kadmium i jord avtar eller øker over tid. Basert på den nåværende kadmiumkonsentrasjonen i jord, mengden tilført kadmium og skjebnen til kadmium i miljøet, er konsentrasjonen i de viktigste mat- og fôrvekstene, i overflatevann og endringen av kadmium i jord over tid blitt estimert. Konsentrasjonen av kadmium i planter som brukes til fôr og mat ble estimert ved bruk av overføringsfaktorer mellom planter og jord.

Eksponeringskonsentrasjon i overflatevann ble beregnet ut fra forventede kadmiumkonsentrasjoner ved utvasking. Fire landbruksregioner i Norge, representert av kommunene Ås, Stange, Time og Melhus, har vært inkludert i vurderingen. De dominerende fôr og/eller matvekstene i de ulike områdene, anbefalt bruk av gjødsel og statistikk over omsetning av kalk, er lagt til grunn for vurderingen. Ved bruk av avløpsslam er både maksimal mengde som kan spres i henhold til dagens regulering (et worstcase scenario), og gjennomsnittlig mengde slam beregnet med utgangspunkt i statistikk, tatt med i vurderingsscenariene. Regionspesifikke parametere som jordegenskaper, årlig nedbør og infiltrasjonshastighet i jord, er faktorer som påvirker skjebnen til og transport av kadmium i miljøet. I denne vurderingen ligger dagens landbrukspraksis og dagens bruk av de mest vanlige mineralgjødseltypene (her i konsentrasjonsområdet 25-50 mg Cd kg-1 P) og målte

kadmiumkonsentrasjoner i tilsvarende mineralgjødseltyper, til grunn. For de andre kadmiumkildene til jordbruksjord (husdyrgjødsel, avløpsslam, kalk og atmosfærisk bidrag), er gjennomsnittlige kadmiumkonsentrasjoner brukt. Dette scenarioet har blitt sammenlignet med bruk av mineralgjødsel med de tre maksimal grenseverdiene (ML) 137,4, 91,6 eller 45,8 mg Cd kg<sup>-1</sup> P mineralgjødsel. Dagens regulering er 100 mg Cd kg<sup>-1</sup> P.

**Bakgrunnskonsentrasjon i jordbruksjord, skjebnen i jord og miljø og forventede miljøkonsentrasjoner (PEC=predicted environmental concentration):** Generelt viser dataene for norsk jordbruksjord lave kadmiumkonsentrasjoner (gjennomsnittskonsentrasjon <0,2 mg kg<sup>-1</sup> tørrstoff, TS). Imidlertid viser områder med alunskifer, f.eks. enkelte områder i Stange i Hedmark fylke, klart høyere verdier, med en maksimalt observert kadmiumkonsentrasjon i jordbruksjord på 3,8 mg Cd kg<sup>-1</sup> tørrstoff (TS) og en gjennomsnittskonsentrasjon på 1,7 mg Cd kg<sup>-1</sup> TS. Gjennomsnittlig kadmiumkonsentrasjon i jordbruksjord i Sørøst-Norge, representert av Ås (0,28 mg kg<sup>-1</sup> TS), var høyere enn i Sørvest-Norge, representert av Time (0,22 mg kg<sup>-1</sup> TS), og i Midt-Norge, representert av Melhus (0,11 mg kg<sup>-1</sup> TS).

Estimert tilført kadmium via mineralsk fosfatgjødsel til jordbruksjord var lavest med grasproduksjon i Time og Melhus (ca. 0,14-0,17 g Cd ha-1 yr<sup>-1</sup>), og høyest med vekstrotasjonen poteter, gulrøtter og korn dyrket på alunskifer i Stange (0,99 g Cd ha-1 yr<sup>-1</sup>). For kornproduksjon i Melhus, Ås og Stange (alunskiferområder) var kadmiumbidraget rundt 0,32-0,42 g Cd ha-1 yr<sup>-1</sup>.

Tap av kadmium fra jord gjennom utvasking og høsting av fôr- og matvekster - og hvor utvasking bidrar langt høyere enn fjerning via vekster - er beregnet til å være rundt 3,5 g Cd ha<sup>-1</sup> yr<sup>-1</sup> på Ås og Melhus, og 18-20 g Cd ha<sup>-1</sup> år<sup>-1</sup> for Time og alunskiferområdene i Stange. Det eksepsjonelt høye estimerte kadmiumtapet i Stange skyldes den høye naturlige bakgrunnskonsentrasjonen, 1,7 mg Cd kg<sup>-1</sup> TS. For Time er dette relatert til høy avrenning, men også til jordegenskaper som gir lavere bindingspotensiale av kadmium i jord enn i de andre områdene som ble vurdert.

Det er generelt akseptert at bindingsegenskapene til kadmium i jord har vært overestimert i tidligere risikovurderinger. Sammenlignet med tidligere vurderinger, er bruk av nedjusterte parametere for bindingsegenskaper også en del av forklaringen på et større tap av kadmium fra jord, og dermed en reduksjon av kadmiumkonsentrasjon i jord over tid.

Tre av de fire områdene som inngår i risikovurderingen, viste en klar estimert nedgang av kadmium i jord over tid, uavhengig av de vurderte ML-verdiene i mineralgjødsel. Det kan imidlertid skje at kadmium i jordbruksareal akkumuleres i områder hvor tilført kadmium er høyere enn tap fra jord. Det gjelder for eksempel Ås, ved bruk av mineralgjødsel med ML-nivå 91,6 mg og 137,4 Cd kg<sup>-1</sup> P og maksimal mengde avløpsslam som er tillatt i henhold til dagens regelverk (et realistisk worst-case scenario). I et 100-års perspektiv er det estimert en økning på henholdsvis 4 og 11 % på Ås. Dette indikerer at på Ås, kan bruk av mineralgjødsel med maksimal Cd-konsentrasjon i forhold til dagens regelverk (100 mg Cd kg<sup>-1</sup> P) sammen med maksimal mengde avløpsslam med en gjennomsnittlig Cd-konsentrasjon

(0.6 mg Cd kg<sup>-1</sup> TS), gi en langsom økning og ikke nedgang av Cd-konsentrasjonen i jord over tid.

I enkelte typer vekster dyrket på alunskiferområder i Stange, er det estimert kadmiumkonsentrasjoner som overstiger grenseverdien for mat, 100 µg Cd kg<sup>-1</sup> friskvekt (FW). Slike høye konsentrasjoner skyldes ikke kadmiumkonsentrasjonen i mineralgjødsel, men den høye bakgrunnskonsentrasjonen av kadmium i alunskiferjord.

Basert på dagens bakgrunnskonsentrasjoner av kadmium i jord, ble kadmiumkonsentrasjonen i overflatevann (PEC<sub>Surface water</sub>) estimert til 0,02  $\mu$ g Cd L<sup>-1</sup> i Melhus, 0,06  $\mu$ g Cd L<sup>-1</sup> i Ås og 0,08 g Cd L<sup>-1</sup> i Time. I alunskiferområdet i Stange, er estimert konsentrasjon 0,24  $\mu$ g Cd L<sup>-1</sup>. Bortsett fra Ås, med bruk av mineralgjødsel med ML 91,6 mg og 137,4 Cd kg<sup>-1</sup> P og tilført maksimal mengde slam, forventes PEC<sub>Surface water</sub> å avta med tiden.

### Risikokarakterisering, miljø:

Risikokarakterisering av jordbaserte planter og dyr og vannlevende organismer, har blitt utført på grunnlag av forventede miljøkonsentrasjoner (PEC) av kadmium i jord og overflatevann, og de forventede null-effektkonsentrasjonene (PNEC) for hver kategori av organismer. Det såkalte risikokarakteriseringsforholdet (RCR), definert som PEC / PNEC, er et kvantitativt uttrykk for risiko. Ved risikokarakteriseringsforhold høyere enn 1 er det sannsynlig at det oppstår uakseptable effekter på organismer. Jo høyere forholdet er, desto mer sannsynlig er det at det kan oppstå uakseptable effekter. Risikokarakteriseringen ga følgende resultater:

<u>Jordlevende organismer:</u> Risikokarakteriseringsforholdene er under 1 for alle scenarier, noe som indikerer ingen risiko for terrestriske organismer ved direkte eksponering for jord. De høyeste risikokarakteriseringsforholdene (0,74) er funnet i Stange, hvor den nåværende bakgrunnen kadmiumkonsentrasjon er 26 % under null-effektkonsentrasjonene i jord.

I de fleste scenarier vil miljøkonsentrasjoner, og dermed også risikokarakteriseringsforholdene, falle over 100 år. Effektene av forskjellig kadmiuminnhold i mineral P-gjødsel er i utgangspunktet ubetydelig, men øker med tiden. Den største økningen er funnet i Melhus, hvor miljøkonsentrasjoner og risikokarakteriseringsforholdt etter 100 år med gjødsel med høyest innhold av kadmium i mineral P er 68 % høyere enn bruk av gjødsel med dagens innhold av kadmium i mineral P i 100 år. Imidlertid er risikokarakteriseringsforholdene fortsatt godt under 1, hvilket indikerer at det ikke er risiko for effekter på terrestriske organismer.

Risikokarakteriseringsforholdene indikerer at det nåværende nivået av kadmium i jord i Stange, medfører en risiko for sekundær forgiftning av landbaserte pattedyr og fugler. Forventede konsentrasjoner viser en nedgang med tiden, men risikokarakteriseringsforholdene er fortsatt over 1 etter 100 år, uavhengig av nivået av kadmium i gjødsel. I scenarier for Ås, Time og Melhus, er alle

risikokarakteriseringsforholdene godt under 1, noe som indikerer at det ikke er risiko for sekundær forgiftning.

<u>Landbruksvekster:</u> Risikokarakteriseringsforholdene er under 1 for alle scenarier, noe som indikerer at det ikke er risiko ved direkte eksponering til jord for landbruksvekster, selv ved det høyeste innholdet av kadmium i gjødsel (137,4 mg Cd kg<sup>-1</sup> P).

Akvatiske organismer: De estimerte konsentrasjonene av kadmium i overflatevann, indikerer at den nåværende bakgrunnskonsentrasjonen av kadmium i jordbruksområder kan utgjøre en risiko for effekter på vannlevende organismer som lever i vannforekomster som mottar dreneringsvann fra jordbruksjord. Dette er særlig tilfellet i Stange, hvor risikokarakteriseringsforholdet basert på nåværende bakgrunnskonsentrasjon er 3. Risikokarakteriseringsforholdet reduseres med tiden ved alle nivåer av kadmium i gjødsel, men er fortsatt over 1 etter 100 år. I Time er risikokarakteriseringsforholdet basert på nåværende bakgrunnskonsentrasjon av kadmium 0,97, men reduseres raskt med tiden. I Ås er det nåværende risikokarakteriseringsforholdet 0,69 og øker med tiden til 0,75 etter 100 år ved maksimal bruk av slam og høyeste ML for kadmium i mineralgjødsel (137,4 mg Cd mg P<sup>-1</sup>) på grunn av akkumulering av kadmium i jord i dette området. Melhus viser laveste risikokarakteriseringsforhold (0,21).

Bidraget fra kadmium fra gjødsel har liten effekt på de beregnede risikokarakteriseringsforholdene på kort sikt, men en økning i kadmiumnivåer i gjødsel (fra anvendt kadmiumkonsentrasjon i vår risikovurdering - 25-50 mg Cd kg<sup>-1</sup> P - til 137,4 mg Cd kg<sup>-1</sup> P), kan etter 100 år forårsake en økning i risikokarakteriseringsforholdene med som så mye som 68 % i Melhus, men bare 5-6 % i Stange, hvor bakgrunnskonsentrasjonen i jord er høy.

<u>Sedimentlevende (bentiske) organismer:</u> I Stange er risikokarakteriseringsforholdene 1 for sedimentlevende organismer med dagens bakgrunnskonsentrasjon, noe som indikerer en risiko. Etter hvert som kadmiumkonsentrasjonene reduseres med tiden, synker risikokarakteriseringsforholdene til under 1 etter 10 og 100 år, noe som indikerer at det ikke er ytterligere risiko for effekter.

For alle scenarier er trendene de samme som beskrevet for overflatevannorganismer. Dette er som forventet, da konsentrasjonene av kadmium i sedimenter er proporsjonale med konsentrasjonene i overflatevann.

### Risikokarakterisering; husdyr og mennesker:

Bortsett fra «worst-case» på Ås, ble det på de andre tre områdene over tid estimert en nedgang i kadmiumkonsentrasjonen i jordbruksjord og planter, og dermed eksponeringskonsentrasjonen via fôr og matvekster.

De tilgjengelige toksikologiske dataene indikerer at doser for hva som kan gi skadelige helseeffekter hos husdyr er langt under estimert eksponering. Men det påpekes at kunnskap om tolerabelt daglig inntak av kadmium hos de forskjellige dyreartene er mangelfull. Kadmium akkumulerer i lever og nyrer, og disse organene fra eldre dyr kan bli brukt som mat, og kadmium fra disse organene kan potensielt bli overfør til mennesker. Det kan tilføyes at den kvantitative sammenhengen mellom kadmium inntak og akkumulering i lever og nyrer er noe usikre.

Ingen av de tre vurderte ML for kadmiumkonsentrasjon i mineralgjødsel som brukes i vurderingen, vil ha noen innvirkning på kostholdseksponeringen for kadmium. I et lengre perspektiv vil kadmiumkonsentrasjonene i planter reduseres på tre av de fire lokalitetene som er vurdert, og dermed vil kostholdseksponeringen for kadmium reduseres. Beregningene indikerer at på Ås med bruk av maksimal mengde tiltatt avløpsslam i henhold til dagens regelverk og maksimale grenseverdier i mineral gjødsel på 91,6 og 137,4 mg Cd kg<sup>-1</sup> P, er det i et 100-års perspektiv henholdsvis 2-3% og 11-12% økning av kadmium i landbruksvekster.

Den nåværende gjennomsnittlige kadmiumeksponeringen overstiger totalt daglig inntak (TWI) for de yngste aldersgruppene, og for de fleste aldersgrupper for høykonsumentene. En reduksjon av kadmiumeksponering er derfor ønskelig, da dette vil redusere risikoen for kadmiumtoksisitet i befolkningen. En reduksjon av kadmiumkonsentrasjonene i de viktigste kildene som korn, poteter og rotgrønnsaker vil redusere eksponering.

VKM har vurdert risikoen for barn som spiser jord. Gitt at denne eksponeringsvei kun skjer i en begrenset periode av livet, og at eksponeringen er lav sammenlignet med eksponeringen via maten, anser VKM at denne eksponeringsveien som lav risiko.

Kadmiumnivåene i drikkevann til husdyr er vanligvis langt under de i för og i beiteplanter, og forventes ikke å bidra betydelig til deres totale kadmiumeksponering.

**Nøkkelord:** VKM, risikovurdering, Vitenskapskomiteen for mat og miljø, Miljødirektoratet, kadmium, mineralgjødsel.

# **Abbreviations**

AF	Assessment factor			
P-AL	Plant-available phosphorus			
BCF	Bioconcentration factor			
BLM	Biotic ligand models			
BMDL	Benchmark dose lower confidence limit			
BW	Body weight			
B2M	Beta-2-microtubulin			
Cd	Cadmium			
Cd <sub>Ex</sub>	Cd excess			
Сфвот	Cd concentration in bottom soil; 50-75 cm soil depth			
Cd <sub>TOP</sub>	Cd concentration in topsoil; 0-25 cm soil			
CDF	Cumulative distribution function			
CEC	Cation exchange capacity			
СР	Cumulative probability			
CN	Curve number			
CSTEE	Scientific Committee on Toxicity, Ecotoxicity and the Environment			
CvM	Cramer-von Mises test			
da	Decare (1000 m <sup>2</sup> )			
DM	Dry matter			
DOC	Dissolved organic carbon			
DW	Dry weight			
DT50	Half-lives			
EC	European Commission			
ECB	European Chemicals Bureau			
ECHA	European Chemicals Agency			
EFSA	European Food Safety Authority			
EQS	Environmental quality standards			
ERA	European Risk Assessment			
ERM	Environmental Resources Management			
FIFRA	Federal insecticide, fungicide, and rodenticide act			
FW	Fresh weight			
ha	Hectare (10000 m <sup>2</sup> )			
HC5	The fifth percentile, with 50% confidence, of a species sensitivity distribution			
IPCS	International Programme on Chemical Safety			
JEFCA	Joint FAO/WHO expert committee on food additives			
K <sub>d</sub> value	Soil and pore-water partitioning or distribution coefficient. Partition coefficient			
	specifically for un-ionized compounds and distribution coefficient for sum un-ionized			
and ionized compounds				
Koc	Partition coefficient			

Kow	Octanol-water partition – experimentally determined			
Кр	Partition coefficient calculated from measured concentrations in European water and sediments			
LCS	Linear concentration shift			
LOEC	Lowest observed effect concentration			
MAC-EQS	Maximum allowable concentrations			
MB	Middle bound			
ML	Maximum limit			
NCA	National Research Council			
NEA	Norwegian Environmental Agency			
NFSA	Norwegian Food Safety Authority			
NGU	Geological survey of Norway (Norges geologiske undersøkelse)			
NOEC	No observed effect concentration			
OPP	Office of pesticide program			
PEC	Predicted environmental concentrations			
PECsw	Predicted environmental concentrations surface water			
PNEC	Predicted no-effect concentration			
PTEs	Potentially toxic elements			
PTMI	Provisional tolerable monthly intake			
PWC	Pesticide water calculator			
RAR	Risk assessment report			
REACH	Registration, Evaluation, Authorisation and Restriction of Chemicals			
RCR	Risk Characterisation Ratio			
SCAHT	Swiss Centre for Applied Human Toxicology			
SCHER	Scientific Committee on Health and Environmental Risks			
SEM	Statistical extrapolation method			
SLV	Livsmedelsverket (National Food Agency in Sweden)			
SOM	Soil organic matter, also often expressed only as organic matter (OM)			
SSB	Statistics Norway (Statistisk sentralbyrå)			
SWCC	Surface water concentration calculator			
TOC	Total organic carbon			
TF	Transfer factor			
TGD	Technical guidance document (issued by European Chemicals Bureau (ECB 2003)			
TWI	Tolerable weekly intake			
UP	Upper intake limit			
US EPA	United States Environmental Protection Agency			
USLE	Universal surface loss equation			
VKM	The Norwegian Scientific Committee for Food and Environment (Vitenskapskomiteen for			
	mat og miljø)			
WC	Water content			
WHO	World Health Organization			

# Background as provided by the Norwegian Food Safety Authority

Phosphorus is an important plant nutrient. The sources for phosphorus in mineral fertiliser contain the toxic metal cadmium. Cadmium is as a result also present as a contaminant in mineral phosphorus fertilisers. However, the amount of cadmium varies greatly between different sources of phosphorus. With repeated fertiliser applications over time, cadmium may accumulate in soil, resulting in negative health and environmental effects.

In Norway, there are two current regulations on marketing of mineral fertiliser products. In the national regulation on marketing of fertilisers and liming materials there is a maximum limit value (ML) of 100 mg Cd kg<sup>-1</sup> phosphorous. The regulation implementing the EU-regulation (EC) No 2003/2003 on EC fertilisers has the same ML as the national regulation. This ML is a result of an adaption text agreed on when the regulation was incorporated into the EEA Agreement, as there is no ML in the EU-regulation.

The European Commission (EC) has suggested a draft template for a new fertiliser regulation covering a wide range of fertilisers and similar products of both organic and inorganic origin. The ML suggested for potential toxic metals vary with product categories. For mineral fertilisers with a total phosphorus content of 5 % phosphorus pentaoxide ( $P_2O_5$ )-equivalent or more, the suggested ML is 60 mg Cd kg<sup>-1</sup>  $P_2O_5$ . After three years, the ML is suggested reduced to 40 mg Cd kg<sup>-1</sup> phosphorus pentoxide. After twelve years, it is suggested reduced to 20 mg Cd kg<sup>-1</sup>  $P_2O_5$ . These MLs will correspond to 137.4 mg Cd kg<sup>-1</sup> phosphorus, 91.6 mg Cd kg<sup>-1</sup> P and 45.8 mg kg<sup>-1</sup> P (mg Cd kg<sup>-1</sup>  $P_2O_5$  = 2.283 x mg Cd kg<sup>-1</sup> P). The suggested levels are still under discussion and it is uncertain whether it is going to be a fixed ML or a ML with downscaling.

The EU institutions have in December 2018 made an agreement on a new ML of Cd at 60 mg Cd  $kg^{-1}$   $P_2O_5$ . This limit will be reviewed 4 years after the date of application (press release Brussels 12.12.2018)

Around year 2000, many countries performed risk assessments of cadmium in mineral phosphorus fertilisers. Such an assessment was also done in Norway (Amundsen et al., 2000). There is now new knowledge about both the fate of cadmium in soil and the health effects of cadmium. Norwegian Food Safety Authority (NFSA) therefore sees the need of a reassessment of the risk.

Amundsen et al. (2000) used agriculture soil data from defined regions as they considered these data to be representative for the future intake of Cd in Norway. If there is a lack of additional soil data, it will be useful for the Norwegian Food Safety Authority if VKM uses the same data as Amundsen et al. (2000), but also in addition to these data we must include

ta from soil types with naturally high cadmium content, such as alum shale soils forts of eastern Norway	und in

# Terms of reference as provided by the Norwegian Food Safety Authority

The Norwegian Food Safety Authority would like VKM to give their opinion on several questions related to cadmium in mineral phosphorus fertiliser.

- What do we know about the levels of cadmium in agricultural soils in Norway today?
- Describe the fate (mobility) of cadmium in agricultural soil and in the local environment, after the application of cadmium-containing mineral phosphorus fertilisers to agricultural land.
- What level of cadmium in agricultural soils would give the risk of negative effects\* on the affected organisms specified in Table A?
- How will application of mineral phosphorus fertilisers with a cadmium content of 137.4, 91.6 or 45.8 mg Cd kg<sup>-1</sup> phosphorus under different crop rotations:
  - affect the levels of cadmium in agricultural soil in Norway in 1, 10 and 100year perspective.
  - affect the risk of negative effects\* on the target organisms specified in Table
     A?
- How will mineral phosphorus fertilisers with a cadmium content of 137.4, 91.6 or 45.8 mg Cd kg<sup>-1</sup> phosphorus affect the dietary exposure to cadmium for Norwegians in general and for subgroups of the population in a 1, 10 and 100-year perspective?

**Table A:** Affected organisms

Affected organism	*Negative effects
<ul> <li>Agricultural plants for food and feed</li> </ul>	Reduced germination/growth and crop
<ul><li>Terrestrial organisms</li><li>Aquatic organisms</li></ul>	Ecotoxicology (environmental risk)
Domestic animals for food production, eating feed and/or grass from fields where mineral fertilisers has been used	Reduced animal health

## **Assessment**

## 1 Introduction

### Occurrence and properties of cadmium

Cadmium is a trace element that occurs naturally in the Earth's crust, where it is found in association with zinc sulphite-based ores, and, to a lesser extent, in lead and copper ores. The content of cadmium (Cd) in soil is due to inputs from both geogenic and anthropogenic sources.

Cadmium forms numerous compounds with other elements and is used in batteries, solders, semiconductors, solar cells, plastic stabilisers, and to plate iron and steel. Salts of Cd show various colours and have been used as pigments in a variety of applications, such as colouring plastics and ceramics. Cadmium can thus enter the environment from various anthropogenic sources, including by-products from zinc-refining, coal combustion, mine wastes, electroplating processes, iron and steel production, pigments, fertilisers, and sewage sludges (NRC, 2005).

Cadmium is a non-nutritive element and has no known physiological effects in plants, animals, or humans. However, it has been shown that Cd may play a role in the enzyme carbonic anhydrase in marine diatoms (Lane et al., 2005).

Cadmium is toxic at very low exposure levels, and has acute and chronic effects on health and the environment. The chemical properties of Cd are similar to those of zinc, which is an essential element, and the two elements interact at many sites in organisms. Cadmium in soil is taken up by plants through the root system and is transferred through the food chain to grazers and carnivores, including humans.

Food is the main source of Cd exposure in the general population; more than 90% of the total intake for non-smokers is via food (EFSA, 2012). Water may also be an important source of exposure. Vegetables, fruits, and nuts are usually low in Cd, but seafoods and certain grains may contain elevated levels (Pond et al., 2004).

### Cadmium in mineral fertilisers

Cadmium is one of the trace elements highly enriched in rock phosphate, which is used for production of mineral fertiliser. An average Cd concentration of 18 mg kg $^{-1}$  in rock phosphate is reported (Altschuler, 1980). The amounts of Cd vary among sources, even in the same deposit (Mar and Okazaki, 2012). Depending on the parent mineral, mining technologies etc, the Cd concentration in commercial mineral fertiliser will vary, e.g., 1-200 mg Cd kg $^{-1}$  P $_2$ O $_5$  (2.3-458 mg Cd kg $^{-1}$  P) (Singh, 1994; Smolders, 2013).

P fertilisers are one of the main sources of Cd in arable soils, and the maximum limit (ML) for Cd in fertiliser is regulated. In 2002, the Scientific Committee for Toxicity, Ecotoxicity and Environment prepared an Opinion on "The Member State Assessments of the Risk to Health and the Environment from Cadmium in Fertilizers" (CSTEE, 2002). Since then, new scientific information has become available and new assessments have been performed (Six and Smolders, 2014; SCHE, 2015; SCAHT, 2015).

As part of the revision of Fertiliser Regulations (EU2003/2003), a ML level of Cd in P fertiliser was proposed in 2016 by EU to facilitate trade of fertilisers between EU Member States, with a goal to reduce accumulation in soil and protect surface waters. The regulation implementing the EU-regulation (EC) No 2003/2003 on EC fertilisers has an ML value of 100 mg Cd kg<sup>-1</sup> phosphorous.

In this risk assessment, VKM answers questions from the NFSA regarding the current Cd concentrations in agricultural soil in Norway, the levels of Cd that would present a risk of negative effects to different target organisms (Table A), how the application of mineral fertilisers with three different ML values of Cd kg<sup>-1</sup> P will affect the concentrations of Cd in soil, and the related risk of negative effects on the target organisms from a 1-year, 10-years, and 100-year perspective. VKM also addresses how application of mineral fertilisers with the different ML values will affect the dietary exposure to Cd for Norwegians in general, and for subgroups of the population, from the same time perspectives.

In addition to mineral phosphorus (P) fertilisers, bio-based fertilisers such as manure, sewage sludge, and liming materials are also sources for Cd transfer to agricultural soil.

### **Bioavailability**

The bioavailability of Cd depends on its chemical form. Cadmium in ionic form and as salts, such as cadmium chloride, have highest bioavailability. Cadmium is partly in free ionic form in fresh water, whereas in foods, including foods of animal origin, it generally exists in a complex with a variety of ligands, including proteins such as metallothionein (NRC, 2005).

The bioavailability of Cd to aquatic organisms decreases with increasing organic carbon and oxygenation. In freshwater, bioavailability also decreases with increasing water hardness, and, in saltwater, with increasing salinities. Thus, toxicity thresholds for aquatic organisms must be interpreted in the context of the characteristics of the water (NRC, 2005). In soil, pH and organic carbon influence Cd solubility, with lower solubility and bioavailability at higher pH and greater organic carbon content.

### Hazard

In order to assess the risk from Cd applied via fertilisers, the highest level of Cd that is expected to cause no toxic effect needs to be established for the environmental compartments exposed; e.g., soil and surface water. These concentration levels are referred to as predicted no-effect concentrations (PNECs). In the present risk assessment, PNECs

from the European risk assessment (ERA) have been used (ECB, 2007). The toxicity of metals in water is modified by abiotic factors, such as pH, dissolved organic carbon (DOC), and water hardness. In order to account for the modifying effect of hardness on toxicity, the PNEC value is adjusted accordingly. As the hardness level of Norwegian surface waters is generally low, and the PNEC for soft water (<40 mg CaCO<sub>3</sub> L<sup>-1</sup>) is used in the aquatic risk assessment.

Earthworms, which ingest soil, are particularly exposed to Cd via this route and bioaccumulation factors of up to 150 (median 15) have been reported for earthworms (ECB, 2007), showing that secondary poisoning within the terrestrial food chain should also be considered in establishment of PNEC for soil (PNEC<sub>soil</sub>). In the risk assessment report (RAR), kidney Cd concentrations of wildlife were used as an indicator of Cd exposure and risk was used to derive a PNEC<sub>soil</sub> that accounts for secondary poisoning, e.g., in wild birds and mammals.

In humans, diet is the main source of Cd exposure in the non-smoking general population. The absorption of Cd is low (3 - 5%), but it is efficiently accumulated in liver and kidney (EFSA, 2009; VKM, 2015). Cadmium also accumulates in bone during growth and remodelling (VKM, 2015). The biological half-life of Cd is long, being 10 to 30 years in healthy humans (EFSA, 2009; VKM, 2012).

Various major organ systems are affected by chronic consumption of foods or water containing high levels of Cd, with the kidneys particularly, but also the liver, primary target organs in most species (NRC, 2005). The toxicity of Cd is partly alleviated by high dietary zinc, iron, and calcium, probably via a complex interaction between these three elements in Cd protection. Cadmium has specific adverse effects on the kidney, and may induce hypertension and microcytic, hypochromic anaemia.

Nephrotoxicity usually results in the initial signs of toxicosis. Damage to proximal tubule cells and interstitial fibrosis in the kidney cortex result in proteinuria, glucosuria, amino aciduria, and polyuria. In the liver, chronic Cd exposure induces histopathological changes, including intralobular fibrosis, cirrhosis, focal mononuclear infiltration, and proliferation of the smooth endoplasmic reticulum. A mild osteomalacia and anaemia have also frequently been observed after chronic Cd exposure.

A Swedish study concluded that there is a statistical relationship between dietary Cd intake and the risk of suffering a fracture, and that there would be a large social benefit to be gained by reducing Cd intake via food (Kemikalieinspektionen, 2012).

The European Food Safety Authority (EFSA) established a tolerable weekly intake (TWI) for Cd based on the increased risk of reduced kidney function in adults following long-term dietary exposure (EFSA, 2009). The TWI was set at 2.5 µg Cd kg<sup>-1</sup> BW per week (EFSA, 2009).

In their risk assessment of dietary Cd exposure in the Norwegian population, VKM concluded that exposure of the Norwegian adult population to Cd is comparable to that of the adult European population (VKM, 2015). Long-term exposure above the TWI from a regular diet is unlikely in Norwegian adults, but additional consumption of foods with a high Cd content, e.g. crab and fish liver, may lead to an exposure exceeding the TWI (VKM, 2015).

### Cadmium exposure

In order to predict the exposure of organisms living in soil, water, and sediments to Cd, and also to humans and livestock animals, the fate of Cd in soil – including runoff, leaching, removal via plant uptake, and transfer to crops and forage – must be known. VKM was thus also asked to describe the fate of Cd in agricultural soil and in the local environment after application of Cd-containing mineral P fertilisers to agricultural land. The questions asked by the NFSA are illustrated in Fig. 1-1.

The concentration of Cd in soil (predicted environmental concentration, PEC<sub>soil</sub>) over time is the difference between input of Cd and loss of Cd. The input of Cd to agricultural soil occurs via mineral fertilisers, bio-based fertilisers, such as manure and sewage sludge, lime products, and atmospheric deposition. Cadmium loss processes from soil include leaching, runoff (erosion), and removal via transfer to crops that are harvested and removed from the field. If the input is higher than the loss, Cd will accumulate in soil over time; conversely, if input is lower than loss, there will be a decrease in soil over time.

Transfer of Cd to crops and forage can be predicted on the basis of predicted environmental concentrations (PEC) in soil and bioaccumulation factors in plants, and further used for predicting human and livestock Cd exposure. Cadmium in surface water might also contributes to human exposure. Leaching of Cd to surface water can be predicted, and the result used to estimate the exposure concentration of Cd in drinking water. For grazing livestock, soil is also a part of their Cd-exposure.

### Risk characterisation

By comparing PEC in soil and surface water with PNEC, the risk of negative effects on soil-dwelling organisms, plants, organisms consuming soil-dwelling organisms and plants, aquatic organisms, and sediment-dwelling organisms, can be evaluated. Correspondingly, by comparing the exposure data with the knowledge on the hazard characterisation, the risk of adverse effects in farm animals and humans can be assessed.



**Fig. 1-1.** An illustration of the elements of this risk assessment: Cd input via mineral P fertiliser (in addition to input from other sources), fate in soil, removal from soil, transfer to crops used for forage, drinking water, and human food.

## 2 Literature search

In recent years, several reports from EFSA and other agencies regarding the risk from Cd in fertilisers and food have been published (Livsmedelsverket, 2017; EFSA, 2012; Rietra et al., 2017; Kemikalieinspektionen, 2012; VKM, 2015; SCAHT, 2015; SCHER, 2015; Finnish Food Safety Authority Evira, 2015). In addition, there are several publications and other reports focusing at modelling and risk evaluation of Cd applied to arable soil via mineral P fertilisers (Smolders 2013; Six and Smolders, 2014). The literature search used in this assessment was focused only towards processes and topics that are directly associated with the possible risks relevant to this risk assessment.

The literature search primarily used the Web of Science, and covered the topics atmospheric deposition, risk of Cd towards livestock, aging of Cd in soil, geogenic versus anthropogenic Cd, plant toxicity, and uptake of Cd in potatoes and carrots. The search terms used, the the number of hits, and refinement hits is shown in Table 2.1.-1.

The approaches and choice of equations and models used for calculations and predicting fate and transfer of Cd, was based on earlier and similar risk assessments performed by VKM (e.g. VKM 2009, VKM 2014) and by Smolders and coworkers (e.g. Six and Smolders, 2014). These are primarily based on technical guidance documents (e.g. ECB, 2003, ECHA, 2016).

**Table 2.1-1.** Summary of literature search topics and terms performed.

Торіс	Data- base	Search terms	Period from	Hits	Refine- ment	Hits refine- ment
Atmosph-eric deposition	Web of Science		1987	29216	1.Cadmium 2. Norway	548 28
Risk livestock	Web of Science	Cadmium/livestock/risk/ assessment		93	,	
Risk livestock	Web of Science	Cadmium/livestock/effe ct/level		75		
Plant toxicity	Google Scholar	cadmium/toxicity/plants /soil/ NOEC	2004- 2018	2380	Original NOECs	2
Uptake in crops	Web of Science	Cadmium AND potato* AND uptake	1987	104		
		Cadmium AND potato* AND peel	1987	15		
		Cadmium AND potato* AND peel AND uptake	1987	7		
		Cadmium AND carrot AND/uptake	1987	46		
		Cadmium AND carrot AND uptake AND peel	1987	2		
		Cadmium AND carrot AND potato AND uptake	1987	14		
		Cadmium AND transfer AND factor AND plant*			Review	11
Uptake of geogenic Cd in plants	Web of Science	Geogenic AND Cd AND health AND plants		17		
Aging of Cd in soils	Google; Web of science	Cd aging AND immobilization	2001	8		
Geogenic vs anthropo-genic Cd	Google	Cd uptake in geogenic AND contaminated soils	1987	12		
Transfer factor	Google	Plant Cd uptake from soil	2001	20		

# 3 Regulations regarding mineral fertilisers and related products in Norway and the EU

### 3.1 Fertiliser

### **3.1.1 Norway**

### Mineral fertiliser

According to Norwegian regulations, the concentration of Cd in mineral fertiliser should not exceed 100 mg Cd per kg phosphorus (mg Cd kg<sup>-1</sup> P) (FOR 2003-07-04 nr 1063).

### Organic fertilisers/Soil amendments

In Norwegian regulations, organic fertilisers/soil amendments are divided into four quality classes based on the content of the potentially toxic elements (PTEs) Cd, Pb, Zn, Cu, Ni, Cr, and Hg (FOR 2003-07-04-951). Maximum level (ML) for As have also been suggested, but are currently not finally accepted. The quality class determines the restrictions regarding application to soil. The ML for Cd in the different quality classes and corresponding maximum application to agricultural soils allowed are given in Table 3.1.1-1.

**Table 3.1.1-1.** Quality classes for organic fertilisers/soil amendments. Maximum levels for Cd in the different quality classes and corresponding maximum application to agricultural soils allowed.

Quality class	Cadmium (mg kg <sup>-1</sup> DW)	Maximum application
0	0.4	No restrictions
I	0.8	40 tonnes DW ha <sup>-1</sup> 10 years <sup>-1</sup>
II	2	20 tonnes DW ha <sup>-1</sup> 10 years <sup>-1</sup>
III	5	No application

### 3.1.2 EU

### Fertiliser

The regulations for EC fertilisers, for use when mineral fertilisers are traded between EU countries (Regulation no 2003/2003), do not include a limit for Cd concentrations in the fertilisers. However, Sweden, Finland and Austria have received exemptions, allowing them to set limits for Cd in EC fertilisers, and the same applies to Iceland and Norway, through the

EEA agreement. These limits are 100 mg Cd kg<sup>-1</sup> P in Sweden and Norway, 50 mg Cd kg<sup>-1</sup> P in Finland and Iceland, and 172 mg Cd kg<sup>-1</sup> P in Austria.

In addition, regulations for national fertilisers (only for national use) exist in many EU countries. The maximum accepted limits for Cd in national fertilisers containing more than 5 %  $P_2O_5$  (2.2 % P) vary from 46 to 206 mg Cd kg<sup>-1</sup> P, with an average of 121 mg Cd kg<sup>-1</sup> P (19 countries) (EC, 2016).

### Sewage sludge

According to the European Council directive on protection of the environment, and, in particular, of the soil, when sewage sludge is used in agriculture, the maximum Cd concentration in sewage sludge that is allowed for application to agricultural soils is 20 to 40 mg Cd kg<sup>-1</sup> dry matter (DW) (EC, 1986). The ML value for the amount of Cd that may be added annually to agricultural land with sewage sludge, based on a 10-year average, is 150 g Cd ha<sup>-1</sup> year<sup>-1</sup>.

### Revision of the EU Fertiliser Regulation 2003/2003

In the draft proposal for the revision of the EU Fertiliser Regulation 2003/2003, it was suggested that the regulations should include limits of PTEs for both mineral and organic fertilisers, and also for soil amendments and growth media. The suggested Cd limit was 137.4 mg Cd kg<sup>-1</sup> P, which was suggested to be reduced to 91.6 mg Cd kg<sup>-1</sup> P after three years and to 45.8 mg Cd kg<sup>-1</sup> P after twelve years. These suggestions were the basis for the present risk assessment. The new rules were approved by the European council on 21.05.2019. Here, the limit for Cd content in "CE marked" phosphate fertilisers is 137.4 mg Cd kg<sup>-1</sup> P, and no reduction of the limit is included. However, when the fertilising product has a lower Cd content than 45.8 mg Cd kg<sup>-1</sup> P, a "low Cd" label may be added.

### 3.2 Soil

#### Norway

In Norway, soils receiving organic fertilisers/soil amendments classified in quality classes I or II should not have Cd concentrations higher than 1 mg Cd kg<sup>-1</sup> DW (FOR 2003-07-04-951).

### **EU**

According to the EU regulations, soils receiving sewage sludge should not have Cd concentration higher than 1-3 mg Cd kg<sup>-1</sup> DW (EC, 1986).

### 3.3 Food and feed

### Norway and EU

Limits for Cd concentrations in the crops included in the selected cases are given in Table 3.3-1. The values are the same for Norway and EU.

**Table 3.3-1.** Limits for Cd concentration in crops included in the selected cases, given in fresh weight (FW).

Crop	Cd (mg kg <sup>-1</sup> FW)
Barley; oat	0.10
Wheat	0.20
Potato (peeled)	0.10
Carrot	0.10
Grass	1.00

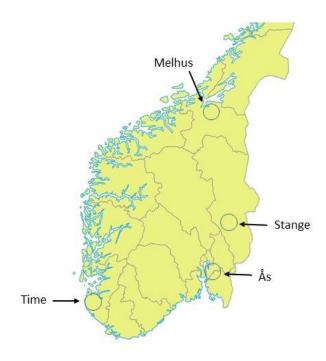
# 4 Description of agricultural regions used in the risk assessment

Soil and climatic conditions, and thereby agricultural practices, vary considerably throughout Norway. In order to account for these regional differences, the risk assessment has been performed for four major agricultural regions (Fig. 4-1):

- 1) Southeastern Norway, with As municipality as a case area,
- 2) Hedmark region, with Stange municipality as a case area for alum shale,
- 3) Southwestern Norway, with Time municipality as a case area,
- 4) Trøndelag (Mid-Norway), with Melhus municipality as a case area.

For Stange and Melhus, two different crop rotations are included, whereas for Time and Ås only one crop rotation is included in the risk assessment analysis.

All measured Cd concentrations in different municipalities and regions are presented in Appendix 1. As the number of soil samples in which Cd concentrations have been measured are limited for each of the municipalities, except for Stange, the Cd concentrations in soil are estimated on the basis of data from the regions and not only the individual municipalities.



**Fig. 4-1.** Regions included in the risk assessment.

### 4.1 Selection of agricultural regions

## 4.1.1 Hedmark county, with Stange municipality as a case area of alum shale

The counties of Hedmark and Oppland both have large areas with alum shale. The alum shale has naturally high content of Cd. The selected municipality, Stange, is located east of Lake Mjøsa in Hedmark county. The area of agricultural land is approximately 9300 ha, of which approximately 8850 ha is tilled land. Agricultural land covers 15 % of the municipality. Grain and oilseed production cover approximately 70 % of the agricultural land, whereas potato- and vegetable production cover approximately 6 % of the agricultural land. Dominant soil types in Stange are loam, sandy loam, and loamy medium sand.

### 4.1.2 Southwestern Norway (Time)

Time municipality is located in the coastal lowland area of Rogaland County in southwestern Norway. The area of agricultural land is approximately 8000 ha, of which approximately 4200 ha is tilled land. Agricultural land covers 37 % of the municipality. Dominant soil types in Time are loamy medium sand and sandy loam.

### 4.1.3 Trøndelag (Mid-Norway, Melhus)

Melhus municipality is located in Gauldalen in southern Trøndelag. The area of agricultural land is approximately 6900 ha, of which approximately 6400 ha is tilled land. Agricultural land covers 10 % of the municipality. Melhus has a wide variation in agronomic practices. Approximately 50 % of the agricultural land is used for grain, 3% for potato, and the rest is used for grass and whole crop silage production. Dominant soil types in Melhus are silt loam, loamy fine sand, clay loam, and loam.

### 4.1.1 Southeastern Norway (Ås)

Ås municipality is located in Akershus county in southeastern Norway. The area of agricultural land is approximately 3950 ha, of which approximately 3880 ha is tilled land. Agricultural land covers 38 % of the municipality. Grain and oilseed production cover approximately 83 % of the agricultural land. Dominant soil types in Ås are silt loam, clay loam, and loam.

### 4.2 Geogenic soil in the selected regions

Surficial deposits (till, gravel, soil) in Norway are largely the result of the latest Quaternary period, with its episodes of glaciation and deglaciation. Most of the agricultural soil was

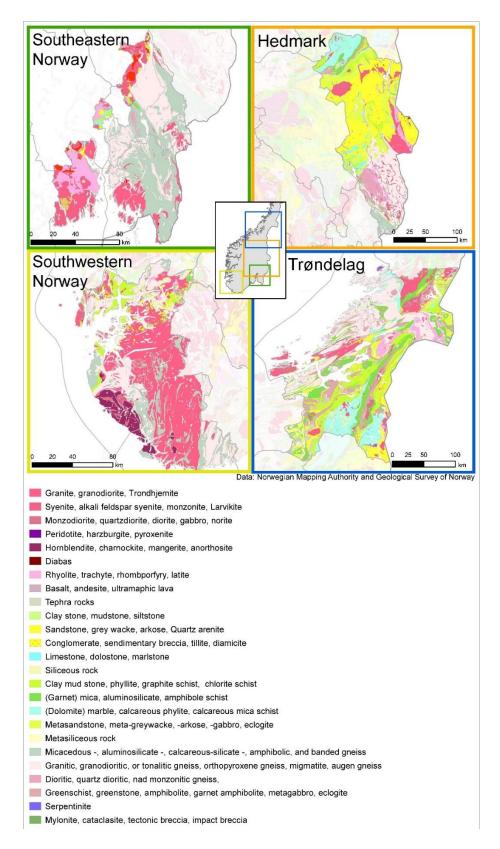
developed on glacial deposits and on postglacial clay-rich marine deposits below the highest coastline. The marine influence on soil can be recognised up to an altitude of ca 200 metres above sea level.

Cadmium can form its own rare minerals, e.g., greenockite (CdS), but is more commonly found incorporated into sphalerite (ZnS), where it often replaces between 0.5-1.5% of the Zn. Traces of Cd also occur in common rock-forming minerals, such as mica and amphibole. While most common rocks contain low concentrations of Cd (0.01-0.25 mg/kg), shale and schist (especially black shales) can show clearly higher values. In Scandinavia, it is common to term Cambrian or early Ordovician black shales as alum shales. These have a wide geographic propagation, not only in Scandinavia but also in, e.g., Central Asia (Christophersen, 2012). In Norway, alum shales are most common in Akershus, Oslo, Oppland (southern part), Buskerud (eastern part), and Hedmark (western part). The Geological survey of Norway (NGU) has, in cooperation with the Norwegian Radiation Protection Authority, made alum shale maps for these counties (http://www.ngu.no/emne/alunskifer-og-radonfare).

Simplified geological maps of the four regions studied, southeastern Norway, Hedmark county, Trøndelag county, and southwestern Norway are shown in Fig. 4.2.1-1, and a more detailed map of Stange municipality in Fig. 4.2.2-1, with identified alum shale areas in Fig. 4.2.2-2.

### 4.2.1 Southeastern (Østfold, Akershus, Vestfold) region

This area is part of the Permo-Carboniferous Oslo Rift, which consists of extrusive and igneous rocks (Permo-Carboniferous) and smaller areas of Cambrian to Silurian sedimentary rocks (Fig. 4.2.1-1). The Oslo Rift is approximately 60 km broad and stretches more than 220 km, from Langesund in the south to the Mjøsa area in the north. It is surrounded by granitoid gneisses of Proterozoic age (Ramberg et al., 2008). The Oslo Rift is dominated by granites, syenites, and monzonites. The soils in this area are glacial deposits of varying thicknesses. Clay, formed in seawater, is common in the bottom of valleys and in the outer parts of Vestfold, which has been below the highest coastline. It is common to find precipitation of iron- and manganese oxides on soils and rocks in streams and lakes in the Oslo Field. These precipitations usually contain PTEs, such as Zn, Pb, Cd, and Mo (Ottesen et al., 2000). The Proterozoic basement surrounding the Oslo Rift is dominated by gneisses, in the north mainly meta-tonalitic gneiss, and granites. In the southeastern part of the region, the soil consists of mainly marine sediments, with elements of glacial deposits. Soil in the northeastern part of the region (Romerike) is dominated by marine sediments, with elements of glaciofluvial deposit. Overbank deposits in the area east of the Oslo fjord show relatively low Zn concentrations compared with the national average, 45 mg/kg HNO<sub>3</sub>-extractable (Ottesen et al., 2000). It is well known that Cd easily substitutes Zn, and low Zn concentrations indicate low Cd concentrations (e.g., Kullerud 1953).



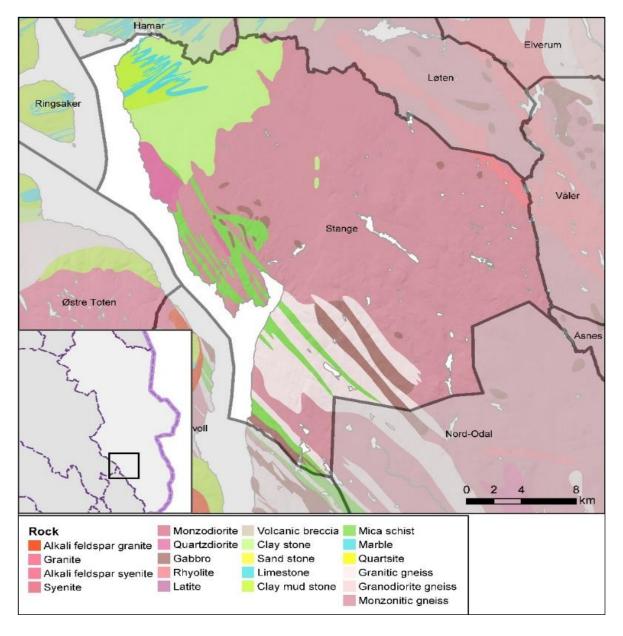
**Fig. 4.2.1-1.** Simplified geological bedrock map of southeastern Norway (Østlandet including Østfold, Akershus, Vestfold county), Hedmark county, southwestern Norway (including Rogaland and Vest-Agder county), and Trøndelag.

#### 4.2.2 Hedmark

In the south, Hedmark comprises the same Proterozoic basement as Østlandet, here mainly dominated by granites, gneisses, and sandstones (Fig. 4.2.1-1). The soil is mainly moraine. Further north, in the area of Lake Mjøsa, the rocks are dominated by slate, limestone, quartzite, and sandstones of mainly Cambrian to Silurian age. The soil consists mainly of continuous thick moraine. Overbank sediments from this area show relatively high Zn concentrations compared with the national average (Ottesen et al., 2000) and therefore high geogenic concentrations of Cd would also be expected. The area north of Lake Mjøsa (Østerdalen) is dominated by thrust nappes composed of Neoproterozoic sandstones and shales. Overbank sediments from this area are generally low in PTEs.

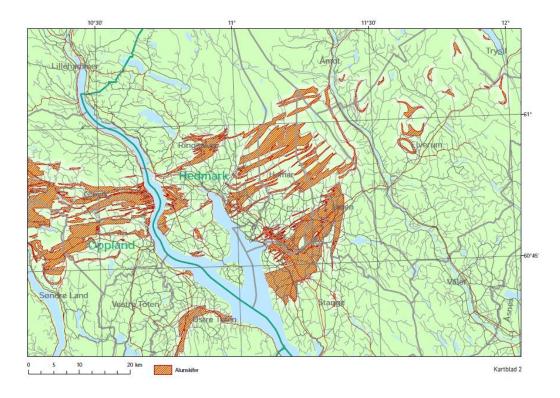
#### Stange municipality

Stange, which is located at the east side of Lake Mjøsa (Fig. 4.2.2-1), is selected as a focus area in Hedmark. The autochthonous rocks in the area are primary augen granite gneiss, tonalite, quartz diorite, mica schist, metasandstone, and quartzite. Stange's largest farms have moraine soil above Cambrian-Ordovician slate and limestone. Although the main part of the fine-grained particles in the moraine are of local origin, most larger rocks, mainly sparagmite sandstone and quartzite, originate from the hills in the north (Dahl et al., 2017). Overbank sediments from Stange show concentrations of Zn twice as high or more than the national average (Ottesen et al., 2000).



**Fig. 4.2.2-1.** Simplified geological bedrock map of Stange municipality. The clay stone, lime stone, and clay mud stone might contain alum shales.

The areas dominated by alum shales in Stange municipality are shown in Fig. 4.2.2-2. The map shows only where alum shales are present as part of the bedrock, close to the surface <a href="http://www.ngu.no/emne/alunskifer-og-radonfare">http://www.ngu.no/emne/alunskifer-og-radonfare</a>).



**Fig. 4.2.2-2.** Alum shale map of the area around Lake Mjøsa showing precautionary areas for radon (http://www.ngu.no/emne/alunskifer-og-radonfare).

## 4.2.3 Southwestern (Rogaland and Vest-Agder county)

The southwestern region consists mainly of the Proterozoic basement gneisses with Caledonian thrust nappes in the west, composed of Precambrian metamorphic and magmatic rocks (Fig. 4.2.1-1). Anorthosites and granitoid gneiss dominate in the south, while mica schists and phyllites (Cambrian and Ordovician), in addition to Precambrian granites, amphibole and charnockite can be found in the northern part of the southwestern region (Fig. 4.2.1-1). In the southern part of the region, analyses of HNO<sub>3</sub>—extract of overbank sediments show Zn concentrations below or close to the national average. In the more northern part of the area, concentrations of HNO<sub>3</sub>—extracted Zn from overbank sediments are significantly above the national average (Ottesen et al., 2000). It is thus also expected that Cd concentrations are higher in this area.

## 4.2.4 Trøndelag (Mid Norway)

Trøndelag (Fig. 4.2.1-1) is the northern-most focus region of this study and comprises Caledonian metamorphic and magmatic rocks, in addition to basement granitoid gneisses that are locally influenced by the Caledonian orogeny. Around the Trondheim fjord, thick layers of clay and other sediments deposited from saltwater predominate. In the remaining area, the main soil type is moraine. Analysis of overbank sediments show low HNO<sub>3</sub>-extractable Zn concentrations in the entire Trøndelag area (Ottesen et al., 2000).

## 5 Common agricultural practice and use of fertilisers

Information about typical crop rotations and recommended fertiliser practices in the different regions was provided by the local agricultural extension service and summarised in Table 5-1. Data on agricultural areas and crops grown in the selected municipalities were obtained from Statistics Norway (SSB) database for the year 2017. In addition, for each of the four counties where the selected municipalities are located, data on the average amount of lime applied in the years 2010-2014 were obtained from the NFSA (NFSA, 2015). Data on average amount of sewage sludge applied in the years 2008 to 2017 were obtained from SSB.

In this risk assessment, we used the given recommended mineral P application from the local agricultural extension service for the different crops in each of the regions (Table 5-1). For cereals, it ranged from 11 to 18 kg P ha<sup>-1</sup> yr<sup>-1</sup> (comparable to 25 to 41 kg  $P_2O_5$  ha<sup>-1</sup> yr<sup>-1</sup>), and for potato and carrot it was, on average, 34 and 24 kg P ha<sup>-1</sup> yr<sup>-1</sup> (comparable to 78 to 55 kg  $P_2O_5$  ha<sup>-1</sup> yr<sup>-1</sup>), respectively. These values are somewhat higher than those used in the risk evaluation by Six and Smolders (2014), who used 21 kg  $P_2O_5$  ha<sup>-1</sup> yr<sup>-1</sup> for cereals and 45 kg  $P_2O_5$  ha<sup>-1</sup> yr<sup>-1</sup> for potato.

**Table 5-1.** An overview of crop rotations, common application of fertilisers, and average application of lime products and sewage sludge in each of the selected case areas; mineral P fertiliser given as kg P ha<sup>-1</sup> yr<sup>-1</sup>, manure as tonnes ha<sup>-1</sup> yr<sup>-1</sup>, lime as kg ha<sup>-1</sup> yr<sup>-1</sup>, and sewage sludge as kg DW ha<sup>-1</sup> yr<sup>-1</sup>. Mineral P fertiliser application rates are average numbers.

Municipality	Ås	Stange	Time	Melhus
County	Akershus	Hedmark	Rogaland	Trøndelag
<b>Crop rotations</b>	Barley-wheat-oat-	Barley-spring	Grass-grass-	Barley-barley-
	barley-wheat	wheat-barley	grass-grass-grass	barley-oat
		Potato-carrot-		Grass-grass-
		grain-potato-		grass-grass-
		grain-grain		grass-whole crop
Applied P via mineral	Barley: 16	Barley: 16/6 <sup>1)</sup>	0	Barley: 13
fertiliser	Oat: 14	Wheat: 18/5 <sup>1)</sup>		Oat: 11
(kg P ha <sup>-1</sup> yr <sup>-1</sup> )	Wheat: 14	Potato: 34		Grass: 6
		Carrot: 24		
Applied	0	Barley and	Grass: 30+20	Grass: 30+20
manure		wheat: 35		(cattle manure)
			(cattle or pig	
(tonnes ha <sup>-1</sup>		(pig manure)	manure)	Whole crop: 35
yr <sup>-1</sup> )				(cattle manure)
Applied lime	130 <sup>3)</sup>	90 <sup>3)</sup>	330 <sup>2, 3)</sup>	220 <sup>3)</sup>
(kg <sup>-1</sup> ha <sup>-1</sup> yr <sup>-1</sup> )				
Applied	55 <sup>4)</sup>	35 <sup>4)</sup>		
sewage				
sludge	4000/2000 <sup>5)</sup>	4000/2000 <sup>5)</sup>		
(kg <sup>-1</sup> ha <sup>-1</sup> yr <sup>-1</sup> )	2)D	and the description of the		2014 (NECA 2015)

<sup>&</sup>lt;sup>1)</sup>Without/with manure; <sup>2)</sup>Presumed spreading on tilled agricultural land; <sup>3)</sup>Average for 2010-2014 (NFSA, 2015); <sup>4)</sup>Average for 2008-2017 (SSB); <sup>5)</sup>Maximum allowed applied, according to regulation for sewage sludge quality classes I and II, respectively (Table 3.1.1-1).

## 5.1.1 Southeastern Norway (Ås)

The usual crop rotation is alternating barley, wheat (both spring and winter wheat), and oats, but with barley and wheat grown more often than oats. The typical chemical fertiliser for grain is Yara compound fertiliser NPK 22-3-10. In addition, wheat is top dressed with mineral N fertiliser.

## 5.1.2 Alum-shale region (Stange)

The usual crop rotation is alternating barley and spring wheat, but with barley grown more often than spring wheat. Manure, if applied, is typically pig manure and is combined with

Yara compound fertiliser NPK 25-2-6 and Opti-NS. Without manure, Yara compound fertiliser NPK 22-3-10 or 20-4-11 is used. A normal crop rotation in which potato and carrot are included is 1: Potato, 2. Carrot, 3. Grain, 4. Potato, 5. Grain, and 6. Grain. For carrot and potato, Yara compound fertiliser NPK 12-4-18 is most commonly used.

#### **5.1.3 Southwestern Norway (Time)**

The usual crop rotation is five years with meadow, after which the meadow is ploughed and reseeded. The grass is usually harvested three times per year, and the total yield is typically 10-13 tonnes DW per ha. Manure is applied in spring and after the first harvest, together with mineral N fertiliser. After the second harvest, only mineral fertiliser is applied.

## 5.1.4 Trøndelag (Mid-Norway, Melhus)

Usual crop rotation in the bottom of the valley is 1.-3. Barley and 4. Oats, whereas on the hillsides, the usual crop rotation is five years with ley, after which the ley is ploughed and reseeded with grain as cover crop and harvested as whole crop for silage. Grain is typically fertilised with Yara compound fertiliser NPK 22-3-10.

Grass is usually harvested twice per year. Manure is applied in spring and after the first harvest, together with mineral fertiliser; Yara compound fertiliser NPK 25-2-6 in spring and mineral N fertiliser after the first harvest. Whole crop is fertilised with manure.

## 6 Fate of cadmium in soil

## **6.1 Influencing parameters**

The fate of contaminants in soil is influenced by their degradation, runoff, and leaching, and evaporation properties. Uptake of contaminants into plants that are harvested and removed from the field also results in loss of Cd from soil and should be included in the fate prediction. A contaminant's physicochemical properties, the soil properties, and climatic conditions will affect these fate processes. Degradation and evaporation are not relevant processes for Cd, and therefore, the important processes to focus on are leaching, runoff, and removal via plant uptake.

#### 6.1.1 Distribution coefficient, Kd

A key parameter for predicting the fate of Cd is its sorption capacity to soil. Sorption processes are represented by the distribution of a compound between the soil (solid phase) and the soil water (water phase), and this ratio is referred to as the distribution coefficient, Kd (typical unit L kg<sup>-1</sup>). For long-term simulations, the Kd is the central and most sensitive parameter. Kd values may be derived from laboratory studies or from measurements in soil *in situ*.

Sorption processes determine the leaching and the runoff rates ( $k_{leaching}$ ), and uptake into and removal via plants ( $k_{plant}$ ). Therefore, the Kd value strongly affects the removal rate from soil and will, for a given input, determine whether Cd will accumulate in soil over time or not. The Kd value will also determine transfer of Cd to surface water, forage and food, and, thus, the exposure concentration to aquatic organisms, and to humans and farm animals via drinking water, crops, and forage.

The Kd value is therefore essential for the risk assessment, and hence selection of the Kd is critical. A high Kd used in simulations over 100 years, will lead to predictions of a much higher accumulation in soils than a lower Kd. Using a Kd that is too high would lead to an underestimation of leaching and of exposure concentrations to aquatic and sediment organisms and to exposure via drinking water. On the other hand, in the long run, uptake into plants, and the resulting exposure of livestock and humans via forage and food, would be overestimated, and vice versa.

Due to the lack of empirical Kd values obtained from studies of Norwegian soil, Kd values were predicted from empirical regressions. The soil parameters that most influence the Kd of Cd are pH and soil organic matter (SOM; commonly expressed only as OM). Thus, both these are frequently applied predictor variables for the Kd of Cd (McBride et al., 1997; Christensen, 1989; Römkens and Salomons, 1998; Sauvé et al., 2000). The Kd is proportional to both parameters and increases with increasing pH and OM (see Appendix III).

In the previous risk assessment of Cd in Norwegian soils (Amundsen et al., 2000), Kd was calculated using the algorithm of McBride et al. (1997). The regression was derived from Cd data measured in 31 soils. This method gives relatively high Kd-values for Norwegian conditions, and a default Kd-value of 500 L kg<sup>-1</sup> was used by VKM (VKM, 2009). Since then, Sauvé et al. (2000) has published a new regression. Sauvé was co-author of the McBride article from 1997, and, in this newer study, a far broader database (830 data points) was used to derive the regression. Smolders (2013) later derived a regression using *in situ* measurements of soil-pore water concentrations of Cd. In contrast with the extraction method used in the earlier studies, *in situ* concentration measurements do not affect the sample pH and are therefore expected to be more realistic (Smolders, 2013). The equation used by Smolders (2013) is based on data from four papers, including Degryse et al. (2003). Kd values calculated according to Smolders (2013) are lower than those of McBride et al (1997), but similar to those calculated by Sauvé et al. (2000).

An evaluation of different Kd regressions, along with the arguments for the selection of the regression, is presented in Appendix III. Based on this evaluation, the regression derived by Smolders (2013) for the Kd of Cd in soil was chosen for the current risk assessment.

#### 6.1.2 pH and SOM

The pH measurements of Norwegian agricultural soils are performed with distilled water  $(pH(H_2O))$ , and for converting to pH measured with calcium chloride  $(pH(CaCl_2))$ , a correction factor is used:

$$pH(CaCl_2) = pH(H_2O)-0.5$$
 (Eq. 1)

The pH of Norwegian agricultural soils is mostly in the acidic range; the mean values for the four municipalities and the different Norwegian regions are all in the range 5.8-6.2 as  $pH(H_2O)$  and 5.3-5.7 as  $pH(CaCl_2)$  (Table A VI-1. Appendix VI) (From NIBIO Soil database). Considerable variations in pH are typical for all the regions, and there are small differences in means over time (Table A VI-1 Appendix VI).

The cold, humid climate in Norway is the main reason for the relatively high content of SOM (Table A VI-2 Appendix VI). The mean content of SOM is highest in Stange/Hedmark (6.7%) and lowest in Time/Rogaland (4.1%). Assuming that SOM is equal to  $1.724 \times 1.724 \times 1.$ 

#### **6.1.3 Precipitation and temperature**

Precipitation and infiltration rate vary considerably in Norway, and, due to the Norwegian climate, precipitation exceeds evaporation in all major agricultural areas. In southeastern Norway, precipitation varies from 300-800 mm annually, but some areas of this region also have extremely low precipitation, being less than 300 mm annually. Southeastern Norway is characterised by relatively high mean temperature, little wind, and low air humidity.

Precipitation is higher in western Norway, most agricultural areas in this region have rainfall in the range of 1200-1500 mm annually. In general, this region is the windiest in Norway and has the highest relative air humidity. In the mid and northern parts of Norway, annual precipitation is relatively high, and the area is characterized by high wind velocities and low temperatures. In the western and northern parts of Norway, the drainage rates are quite high and the typical drainage rates for the agricultural areas are generally higher than those in southeastern Norway. These climatic variations are the reason why the amount of precipitation that infiltrates the soil also varies throughout the country.

In this type of risk assessment, soil infiltration, which is commonly called precipitation excess, is often given as a fraction of the total precipitation. In the calculation of soil concentration in the risk assessment of sewage sludge (VKM, 2009), a mean precipitation excess of 0.25 was used, which is identical to the default value suggested in the Technical guidance document (TGD) (ECB, 2003). In the risk evaluation of fate and effects in the Zn and Cu in pig and poultry production (VKM, 2014), 0.4 was used for Åsnes in Hedmark and 0.7 for Klepp in Rogaland and for Melhus in Trøndelag. In the present risk assessment, the same precipitation excess has been used for locations close to those places used by VKM (2014) (see Table 8.1-1).

Using values for the precipitation excess that are too low will result in predicted soil accumulation being too high and the predicted transfer to nearby water recipients being too low. If precipitation excess values are too high, the predicted soil accumulation results will be too low and the predicted transfer to water recipients too high.

## 6.2 Removal processes from soil

## 6.2.1 Leaching

Different hydrological processes may transport water that falls on soil surface and thus influence leaching patterns of Cd. Precipitation may infiltrate the upper soil and percolate through subsoil to lower layers (leaching) or flow laterally as overland flow (runoff) on the soil surface.

Water infiltrating soils may flow downwards through soil matrix, and if the water volume are larger than soil matrix is able to transport and macropores occur, water may be transported also in these. Macropores are pores that are significantly larger than those resulting from simple packing of elementary particles (Bouma, 1982), and may occur due to soil structural cracks, faunal activity or root channels. Whereas clay rich soils are exposed to cracking, clay is also characterized by low hydraulic conductivity, and such structured soils are often characterized by rapid fluxes of water in large pores and cracks. Macropore flow and runoff represent bypass flow with respect to leaching and may affect the transport of solutes (eg Cd) because these processes involve less interaction between the water and soil as compared to leaching. Runoff may also include erosion and transport of Cd that is bound to particles. However, these bypass transport processes have not been considered in the

calculation of loss of Cd from soil. Instead it has been assumed that all precipitation excess water leaches through top soil, via deeper soil layers, to ground water or to drainage systems, and, further, to nearby surface water recipients. The leaching rate from soil ( $k_{leaching}$ ), according to equation 2, is commonly included and calculated in risk assessments (ECHA, 2016).

$$k_{leaching} = \frac{Finf_{soil} \cdot RAINrate}{K_{soil-water} \cdot DEPTH_{soil}}$$
 (Eq. 2)

Where

Finf<sub>soil</sub> – fraction of rain-water that infiltrates into soil

RAIN<sub>rate</sub> – rate of wet precipitation [m·yr<sup>-1</sup>]

K<sub>soil-water</sub> – soil-water partitioning coefficient [m<sup>3</sup>·m<sup>3</sup>]

DEPTH<sub>soil</sub> – mixing depth of soil [m]

k<sub>leaching</sub> – first order rate constant for leaching from soil layer [yr<sup>-1</sup>]

The soil-water partitioning coefficient,  $K_{\text{soil-water}}$ , is used in calculating the predicted environmental concentration (PEC) in soil, and is calculated using equation 3.

$$K_{soil-water} = Fair_{soil} \cdot K_{air-water} + Fwater_{soil} + Fsolid_{soil} \cdot \frac{Kd_{soil}}{1000} \cdot RHO_{soil}$$
 (Eq. 3)

Where

Fwater<sub>soil</sub> = volume fraction of water in soil compartment [ $m^3 \cdot m^{-3}$ ] (0.2 TGD in ECB, 2003)

Fsolid<sub>soil</sub> = volume fraction of solid in soil compartment [m<sup>3</sup>·m<sup>-3</sup>] (0.6 TGD in ECB, 2003)

Fair<sub>soil</sub> = volume fraction of air in soil compartment  $[m^3 \cdot m^{-3}]$  (0.2 TGD in ECB, 2003)

 $RHO_{soil} = density of the solid phase [kg·m<sup>-3</sup>]$ 

Kd<sub>soil</sub> = solids-water partition coefficient in soil [I·kg<sup>-3</sup>]

 $K_{air-water} = air-water partitioning coefficient [m<sup>3</sup>·m<sup>-3</sup>]$ 

 $K_{\text{soil-water}} = \text{soil-water partitioning coefficient } [\text{m}^3 \cdot \text{m}^{-3}]$ 

Loss of Cd from soil via water transport, e.g. given as g ha<sup>-1</sup> year<sup>-1</sup>, can be predicted based on the leaching rate,  $k_{leaching}$  (equation 2), Cd concentration in soil and soil volume.

Six and Smolders (2014) had another approach, predicting pore-water concentration of Cd (based on Kd) and precipitation excess for estimating loss of Cd per ha per year (equation 4).

$$C_{leaching} = 10000 \cdot F \cdot C_{solution}$$
 (Eq. 4)

Where

F = precipitation excess [m yr<sup>-1</sup>]

C<sub>solution</sub> = Cd concentration in soil solution or pore water [mg L<sup>-1</sup>]

C<sub>leaching</sub> = leaching of Cd per ha per year [g ha<sup>-1</sup> yr<sup>-1</sup>]

In this risk assessment, both these approaches have been evaluated.

## 6.2.2 Removal via uptake in harvested plants

In this risk assessment, uptake of Cd by plants plays a role both in the removal of Cd from the soil (estimate of annual loss of Cd from soil via harvested plants) and in transfer of Cd to the food chain (exposure to humans and farm animals). In this section, removal of Cd from soil via harvest of crops is addressed. If plant biomass is not harvested and transported from the field, then removal via plants would be less than calculated by the plant removal rate,  $k_{plant-removal}$  (year-1), according to equation 5.

$$k_{plant-removal} = \left(\frac{CP \cdot C_{crop}}{DEPTH_{soil} \cdot RHO_{soil} \cdot C_{soil}}\right)$$
 (Eq. 5)

#### Where

 $k_{plant-removal} = plant removal rate [yr^{-1}]$ 

CP = crop production [kg DW·m<sup>-2</sup>·yr<sup>-1</sup>]

 $C_{crop} = concentration in crop [mg \cdot kg^{-1} \cdot DW]$ 

DEPTH<sub>soil</sub> – soil depth [m]

RHO<sub>soil</sub> – bulk density of soil [kg·m<sup>3</sup>]

C<sub>soil</sub> – concentration in soil [mg·kg<sup>-1</sup> DW]

**Table 6.2.2-1**. Crop production (crop yield) for different crops produced in the selected regions, given in kg FW biomass ha<sup>-1</sup> yr<sup>-1</sup>. Data provided by the local agricultural extension service. For prediction of  $k_{plant-removal}$ , FW was calculated to DW using the water content of different crops, as presented in Table 7.2-1.

Crops	Location	Crop yield (tonnes yield FW ha <sup>-1</sup> yr <sup>-1</sup> )
Wheat	Ås	6.8
Barley	Ås	5.8
Oat	Ås	5.2
Wheat	Stange	5.6
Barley	Stange	5.1
Carrot	Stange	40
Potato	Stange	30
Barley	Melhus	3.8
Oat	Melhus	3.6
Grass	Melhus	30
Grass	Time	57.5

The concentrations in agricultural crops,  $C_{crop}$ , are calculated using constant transfer factors (TF; mg Cd per kg plant to mg Cd per kg soil); i.e. plant concentrations are proportional to soil concentrations (Table 7.2-1). This approach is based on the assumption that the concentrations of trace metals in plants or different parts of the plant (stem, leaf, grain etc.) are proportional to the total concentration in soil. The TFs vary for different plant species. There is considerable variation in published TFs (often also named bioconcentration or bioaccumulation factors (BCFs)). The basis for selection of TFs used in this risk assessment is detailed in section 7.1.4 and 7.2, and Appendix IV.

$$Cplant_{potato,cereal,gras} = Csoil \cdot TF_{potato,cereal,gras}$$
 (Eq. 6)

Where

Cplant<sub>potato, cereal, grass</sub> = concentration of Cd in potato, cereal, grass [mg kg<sup>-1</sup> DW] Csoil = total concentration of Cd in soil [mg kg<sup>-1</sup> DW] TF<sub>potato, cereal, grass</sub> = crop-specific TF for uptake of Cd from soils into plants [mg Cd kg<sup>-1</sup> DW plant to mg Cd kg<sup>-1</sup> DW soil]

#### 6.2.3 Aging processes

The potential mobility and availability of Cd depend on its total concentration in the soils, in soil solution, and in exchangeable forms. Retention and release reactions of solute with different components of the soil matrix govern the chemical behaviour of Cd. For Cd recently added to soils, partitioning between the soil solution and the solid phase gradually changes with time, until it reaches a state of pseudo-equilibrium. The rate at which this is attained is not only a time-dependent process, but is also governed by the nature of the element and soil properties (e.g., pH, organic matter, and clay content).

Prediction of potential mobility, and the bioavailability of Cd and other trace elements requires both chemical and biological methods, especially to quantify the fraction of Cd available for biological uptake. Chemical surrogates for assessing bioavailability have been sought due to their relative simplicity and rapidity in comparison with biological methods (Singh, 2007). Retention of Cd can be detected with high sensitivity by measuring the change in the fraction of the total concentration that is isotopically exchangeable, the ratio of the so-called E value to the total concentration (Smolders et al., 1999). This fraction is conceptually identical to the fraction of Cd that is available as freshly applied metal. In a greenhouse study with 28 soils that were either spiked with a mixture of metals, including Cd, and aged > 2 years on the field and in corresponding soils that were freshly spiked with identical total metal concentrations, Buekers (2007) found that aging significantly reduced shoot Cd concentrations in 3 of 19 soils. The change in Cd concentration in shoots was significantly associated with corresponding changes in isotopically exchangeable concentrations (E values) for Cd. The aging effect predicted that Cd decreases proportionally with decreasing E values during aging. Multivariate analysis showed that aging effects on Cd were significant when using total Cd concentration, and that the E values explained these aging effects; Cd concentrations decreased proportionally with decreasing E values during

aging. However, compared to other processes that affect bioavailability, the effect of ageing is relatively small. Similarly, in a growth chamber study, Hamon et al. (1998) reported that in soils in which Cd application had been stopped after 27 years of mineral fertiliser application, 40% of the Cd added over time was in plant-available form. However, in soil where Cd application was continuing (47 years later) about 72% of the Cd was in plant-available form. Using a model, and assuming a constant rate k of fixation of Cd added to soil, the amount of Cd that is available (labile Cd) at time "t" could be estimated. The model showed that Cd was fixed in the soil at a rate of 1-1.5 % of the total Cd added per year. The decrease in availability in the continuous and rundown plots indicates that stabilisation processes occurring over time decreased the availability of Cd added to the soil. This stabilisation may provide a partial explanation for the lack of a consistent trend in increasing Cd availability over time with repeated application of Cd in mineral fertiliser.

From a long-term (1992 to 2002) experiment in Norway, Chaudhary et al. (2011) found an increase in E values with time, but no significant pH- or Cd application-dependent effects on E values could be identified. This indicates that Cd added anthropogenically (fertilisers and probably lime) remained predominantly as exchangeable Cd. From the same long-term experiment, Singh et al., (2017) found that the extractable Cd in the soil decreased from 1992 to 2002, but the decrease was not statistically significant, and was more influenced by soil pH. Thus, the ageing effect apparently has less influence than Cd added by fertiliser and/or lime.

The above-cited evidence indicates that metals, including Cd, are adsorbed to the soil matrix over time. Nevertheless, it is difficult to make quantitative estimates of the rate of fixation as it is affected by the nature of the metals and the soil properties. A value of 0.5 to 1% fixation of Cd per year was proposed by Hamon et al. (1998). However, as more recent studies do not support this, but indicate that aging apparently does not seem to play a major role when Cd is added by fertiliser and/or lime, such in this risk assessment, aging was not taken into account here in the current evaluation.

# 7 Hazard identification and characterisation

## 7.1 Cadmium in soil: background, input, and loss

This chapter covers Cd concentrations in background agricultural soil, Cd content of fertilisers and liming products, and atmospheric deposition of Cd, along with the selection of the different input parameters.

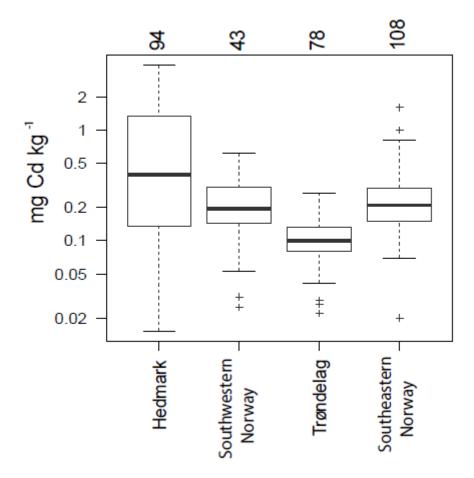
#### 7.1.1 Soil background

There are generally few data available on Cd content in Norwegian soil. An overview of number of samples from each municipality in the different regions is given in Appendix I. Table 7.1.1-1 gives the minimum, mean, median, and maximum Cd concentrations in each region based on all the data presented in Appendix I and illustrated in Fig. 7.1.1-1.

Alum shale containing naturally high levels of Cd is of special importance in this risk assessment. Stange municipality in Hedmark has large agricultural areas with alum shale, and, due to the interest in Cd concentrations in this soil, there is a high number of sample locations from this area. Sandefjord municipality in the southeastern region also has more sample locations than most other municipalities. Results from these two municipalities might therefore have a skewing effect on the statistics in their regions, and this is further discussed in Appendix I.

**Table 7.1.1-1.** Cadmium concentrations in background agricultural soil for the selected focus municipalities. Mean values of soil Cd concentrations (mg kg<sup>-1</sup> DW) are used to calculate PEC<sub>soil</sub>.

County	Southeastern	Hedmark		Hedmark		Southwestern	Trøndelag
	Norway Melhus	Stange	Whole	Norway Time	Melhus		
Municipality	Memus	region		Time	Pielius		
Soil concentration (	mg Cd kg <sup>-1</sup> DW)						
MIN	0.020	0.39	0.015	0.025	0.022		
MEAN	0.282	1.70	0.861	0.224	0.108		
MEDIAN	0.211	1.70	0.400	0.194	0.100		
MAX	1.800	3.84	3.84	0.614	0.270		



**Fig. 7.1.1-1.** Boxplots of all of all data from four Norwegian regions, compiled from the dataset in Table 7.1.1-1., presented as mg Cd kg<sup>-1</sup> DW. The number of samples in each region is given on the top axis. Note the logarithmic scale of the y-axis.

#### 7.1.2 Input sources of Cd to soil

#### 7.1.2.1 Cd content in applied fertilisers and lime products

Input of Cd to soil through agricultural management practices includes use of mineral P fertiliser, manure, sewage sludge, lime, and biowaste. The source of Cd in mineral fertilisers is Cd in rock phosphate used for fertiliser production. In a former report (Amundsen et al., 2000), input of Cd to soil through purchased feed concentrates was used instead of Cd input with manure, because some of the Cd in manure is part of an internal Cd cycle. Here, we used the amount of Cd in manure as input in order to account for leaching risk of applied Cd. Increasing amounts of biowaste are being applied in agriculture, but the amounts are still small compared with other sources of applied Cd. Therefore, Cd input to soil with biowaste is not included here.

#### 7.1.2.1.1 Mineral P fertiliser

According to the fertiliser recommendations (NIBIO fertiliser handbook¹), the amount of applied P should be adjusted according to soil P status, and P fertiliser should not be used when soil P status is very high (P-AL >14). Here, recommended P fertiliser usage is used for the calculating Cd input to soil with mineral P fertiliser without adjustment for soil P status. Different mineral fertilisers are used for different crops, and, by combination with manure, mineral fertilisers with lower P concentration are used. The concentrations of Cd per kg P in the different types of mineral fertiliser that are commonly used in the case areas are presented in Table 7.1.2.1.1-1. NFSA conducted a survey of Cd content in mineral fertiliser products. The results varied from <2 mg Cd kg⁻¹ P up to 160 mg Cd kg⁻¹ P (n=48). Two products had a higher Cd content than the ML value, and the products were quickly withdrawn from sale. Applied amounts of mineral fertilisers in the case areas are given in Table 5-1.

The typical chemical fertiliser for grain is Yara compound fertiliser NPK 22-3-10 (NFSA, 2018). However, both Yara compound fertilisers NPK 22-3-10 and 20-4-11 are used at Stange.

**Table 7.1.2.1.1-1.** Concentrations of Cd per kg P in the different types of mineral fertiliser (NFSA, 2017), and crops for which they are commonly used.

Fertiliser NPK	mg Cd kg <sup>-1</sup> P	Crop
Yara 22-3-10	25	Grain
Yara 20-4-11	84	Grain
Yara 25-2-6	27	Grain, grass
Yara 12-4-18	50	Potato, carrot

Yara compound fertilisers NPK 22-3-10 and NPK 25-2-6 are produced in Porsgrunn, whereas NPK 12-4-18 and NPK 20-4-11 are produced in Glomfjord, and this might explain the difference in Cd concentrations between the different fertilisers (NFSA, 2017). In the calculations, the Cd concentrations of Yara NPK 22-3-10 and NPK 25-2-6 were used for grain and grass (the most common use in these scenarios), whereas NPK 12-4-8 was used for potato and carrot.

#### 7.1.2.1.2 Manure

Cadmium concentrations in manure presented in Daugstad et al. (2012) show large variations. The average Cd concentration in DW, with variation for different manure types and concentrations in manure with a normalized DW content are presented in Table 7.1.2.1.2-1. The amounts of manure applied in the case areas are given in Table 5-1.

<sup>&</sup>lt;sup>1</sup> https://www.nibio.no/tema/jord/gjodslingshandbok?locationfilter=true (in Norwegian)

**Table 7.1.2.1.2-1.** Average (variation) Cd concentration in DW in different manure types, and Cd concentrations in manure with normalized DW content (Daugstad et al., 2012).

	mg Cd kg <sup>-1</sup> DW	mg Cd tonnes <sup>-1</sup> FW
Cattle manure	0.13 (0.08-0.18)	7.8 (6 % DW)
Pig manure	0.27 (0.12-0.44)	10.8 (4 % DW)
Chicken manure	0.16 (0.08-0.24)	96 (60 % DW)

In the calculations, the average Cd concentrations in cattle and pig manure are used for Time.

#### 7.1.2.1.3 Sewage sludge

Between 2010-2015, the yearly average Cd concentration in sewage sludge was 0.6 mg Cd  $\rm kg^{-1}$  DW (data obtained from SSB database). The average for 2016 was somewhat lower, 0.5 mg Cd  $\rm kg^{-1}$  DW (Berge and Sæther, 2017). Here, we have chosen to use 0.6 mg Cd  $\rm kg^{-1}$  DW. The average applications of sewage sludge in Stange and Ås for the years 2008-2017 were 35 kg and 55 DW  $\rm ha^{-1}\,yr^{-1}$ , respectively (Table 5-1).

The amount of sewage sludge allowed for application is regulated according to the heavy metal concentration. The limit for Cd concentrations in quality classes I and II are 0.8 and 2.0 mg Cd kg<sup>-1</sup> DW, respectively (FOR 2003-07-04 nr 951), that is, the average Cd concentration in sewage sludge is within quality class I. The maximum sewage sludge application allowed according to current regulations is 20 tonnes DW ha<sup>-1</sup>10 yr<sup>-1</sup> for sludge in quality class II and 40 tonnes DW ha<sup>-1</sup>10 yr<sup>-1</sup> for sludge in quality class I. The commonly applied amount is 20 tonnes DW ha<sup>-1</sup>10 yr<sup>-1</sup>, and this is usually applied in a single dose that results in 12 000 mg Cd ha<sup>-1</sup> in the year of application, assuming 0.6 mg Cd kg<sup>-1</sup> DW.

In Ås and Stange, Cd contribution via application of sewage sludge is included. We have used both the maximum allowed amount and the amount based on numbers from SSB, in the calculation.

#### 7.1.2.1.4 Lime products

The average Cd concentration in 16 different Norwegian lime products of various origins was 0.06 mg Cd kg<sup>-1</sup> lime (Erstad, 1992). The minimum and maximum values were <0.01 and 0.33 mg Cd kg<sup>-1</sup>, respectively. The amounts of lime applied in the case areas are given in Table 5-1.

#### 7.1.2.2 Atmospheric deposition of anthropogenic Cd

**Trends of atmospheric deposition of Cd.** Investigations around smelters have repeatedly shown that Cd concentrations decrease exponentially with distance from source (e.g., Bonham-Carter and Mc Martin, 1997). The quantification of the contribution of diffuse Cd

emissions to the Cd concentration in soil is a matter of debate, due to the high natural background variability (orders of magnitude) of Cd concentrations in soil.

The most recent estimate of Cd deposition to European agricultural soil is provided by Six and Smolders (2014). These authors estimated an input via atmospheric deposition at the European scale of 0.35 g Cd ha<sup>-1</sup> yr<sup>-1</sup>, with a declining tendency. Nationwide moss surveys performed in Norway regularly since 1977 show that the same declining tendency also applies to Norway (Fig. A II-1) (Steinnes et al., 2016; Berg et al., 1994; Berg et al., 1995, Appendix II). The atmospheric contribution, as measured by the air- and precipitation monitoring network in Norway (Bohlin-Nizzetto et al., 2018), varies from between 0.05 and 0.36 g Cd ha<sup>-1</sup> yr<sup>-1</sup>, with Birkenes in the south and Svanvik in the north showing the highest concentrations (Appendix II).

Biospheric Cd. Cadmium has a very high affinity to the biosphere. Marine bird guano is, for example, known as a possible source of extreme Cd concentrations in soil (e.g., Garrett et al., 2008, 2010), indicating that Cd accumulates in the food chain. Many plants strongly enrich Cd in their foliage, partly to defend themselves against grazing animals (e.g., Gough et al., 2013). Plants growing at the same site and under the same environmental conditions show huge differences in Cd concentrations. This strongly suggests that bio-productivity and plant population affect the Cd concentrations in the local organic topsoil, which, to a very substantial part, consists of decaying plant material (Flem et al., 2018; Reimann et al., 2018a, b; Andersson et al., 2018). Cadmium is the element that is most often seen enriched in forest floor O-horizon soil when compared with the underlying mineral soil C-horizon, independent of where the samples are taken (e.g., Reimann et al., 2009, 2015). This is mainly due to a natural process (the 'plant pump') translocating Cd from mineral soil to organic surface soil, first described by Goldschmidt (1937). Goldschmidt (1937) observed that germanium (Ge), with a crustal abundance of a few mg kg<sup>-1</sup>, can be accumulated in certain coal ashes to a level of 1%. This not only relates to Ge, as several studies have shown that silver (Aq), mercury (Hg), Cd, and even gold (Au), all tend to accumulate strongly in the soil O-horizon (Reimann et al. 2007a, 2015). The bio-activity is often not taken into consideration when calculating 'atmospheric deposition'. Additionally, the atmosphere will always reflect local dust sources, as well as long-range atmospheric input from anthropogenic sources. This was well demonstrated by a survey that sampled mineral soil C-horizon, organic soil O-horizon, and terrestrial moss (Hylocomium splendens) along a 100-km transect, over two mineral deposits in southern Norway; the Nordli Mo deposit, with surface exposure of molybdenite (MoS<sub>2</sub>), and a sandstone-hosted Pb-mineralization with surface exposures of sphalerite (Flem et al., 2018). Results demonstrated that moss reacts strongly to the presence of local Pb mineralization (Flem et al., 2018), as local 'dust' overprints any other Pb sources.

**New methodology for predicting diffuse atmospheric deposition.** Fabian et al. (2017) presented a new method for detecting and quantifying the *diffuse* external input of an element to soil at the continental to regional scale, by comparing its statistical distribution in top soil against that of bottom soil. Diffuse contamination, by definition, generally contributes the same Cd flux everywhere in the same area. In addition, local source

pollutants can vary in the area. This method has been used by Reimann et al. (2019) to estimate diffuse contamination with Cd using eight regional-to-continental-scale datasets, including the Baltic Soil Survey data (Reimann et al., 2003). The Baltic Soil Survey provides a database of more than 60 chemical elements in agricultural soils from an area of 1,800 000 km<sup>2</sup>, including ten northern European countries (western Belarus, Estonia, Finland, northern Germany, Latvia, Lithuania, Norway, Poland, north-western Russia, and Sweden) (Reimann et al., 2003). Cumulative probability (CP) plots for Cd in top soil (0-25cm) and bottom soil (50-75cm) from the Baltic Soil Survey, are shown in Figure 7.1.2.2-1. In order to estimate contamination based on comparison of different sample media, the effects of element dilution and up-concentration have to be considered. Such effects occur through soil development processes (e.g., decomposition of organic material and weathering) that either add or remove elements from the soil layer. The element concentration thereby changes from y to y/(1+x), leading to a linear concentration shift (LCS) of the cumulative distribution function (CDF) in the CP-diagram (Fabian et al., 2017). The observed LCS between top and bottom soil reflects a natural process, leading to a general Cd enrichment in more organic surface soil that cannot be reasonably explained via external input. In contrast, the steepening of the distribution function of the topsoil samples at the lower concentration range is due to input from external sources.

Applying the method given by Fabian et al. (2017), comparing the CDFs of top soil (0-25cm) and bottom soil (50-75cm) from the Baltic Soil Survey according to the model equation:

$$Cd_{TOP} = a Cd_{BOT} + Cd_{EX}$$
 (Eq. 7)

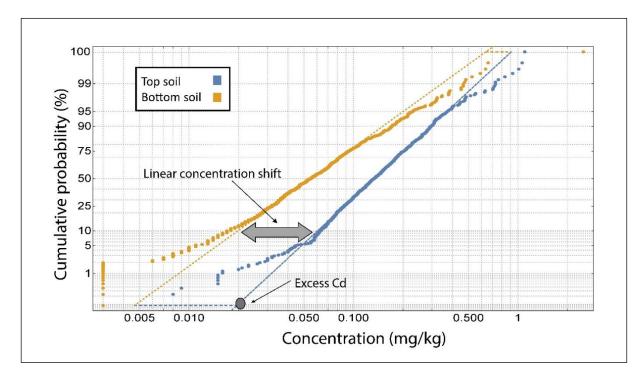
Where:

 $Cd_{TOP}$ =Cd concentration in top soil (0-25 cm) [mg kg<sup>-1</sup> DW]  $Cd_{BOT}$ = Cd concentration in subsoil (50-75 cm) [mg kg<sup>-1</sup> DW]  $Cd_{EX}$  = excess Cd, representing diffuse contamination a= a constant describing the Linear Concentration Shift (LCS)

can be used to determine the optimal values of the LCS, the constant a, and  $Cd_{EX}$ , such that the p-value of the Cramer-von Mises (CvM) test (Cramér, 1928; Mises, 1928), which quantifies the quality of this best fit, is maximal for the resulting CDFs. If CvM>0.05, the assumption that the above simple model is essentially correct on a 5% level cannot be rejected. For the Baltic Soil Survey data, with a=1.87 (LCS) and  $Cd_{EX}=0.028$  mg/kg, results in a test value of CvM=0.4, indicating that both data sets are drawn from the same probability distribution (Fig. 7.1.2.2-1, Reimann et al., 2019).

The data from the Baltic Soil Survey thereby provides a quantitative estimate of the anthropogenic input of Cd to agricultural soil via diffuse contamination in the order of 0.028 mg kg<sup>-1</sup> at the Northern-European scale. Local or regional contamination can substantially add to this value in some areas. When calculating the total upper limit of the total Cd influx (over hundreds of years) to agricultural soil per ha and year, the total mass of excess Cd has to be related to the volume (V) sampled. Using a bulk dry soil density of 1200 kg/m³ and a soil depth

of 25 cm (which was the sampling depth of topsoil at the Baltic Soil Survey), this will correspond to 3000 tonnes soil ha<sup>-1</sup>. Assuming a 200-year period of Cd influx, the yearly Cd diffuse flux to soil surface can then be calculated to be 0.42 g ha<sup>-1</sup> yr<sup>-1</sup>. This value is dependent on an assumed plough depth, time of cultivation, and soil density, which is highly dependent on organic content. The calculation from the upper limit of total influx of 0.42 g Cd ha<sup>-1</sup> yr<sup>-1</sup> influx to agricultural soil is thus only an estimate of the order of magnitude. This estimate of Cd influx is comparable to that suggested by Six and Smolders (2014) for Europe, and to precipitation data given by Bohlin-Nizzetto et al., (2018) for Norway. However, it also includes local addition of Cd through the use of fertilisers. Using a soil depth of 20 cm, as recommended in a technical guidance document from the European Chemicals Agency (ECHA, 2016), as representing the ploughing depth (this range has a high root density of crops), the upper limit of influx is estimated to be 0.34 g Cd ha<sup>-1</sup> yr<sup>-1</sup>. This estimate is also in good agreement with the (measured) wet atmospheric deposition in Denmark of 0.30 g ha<sup>-1</sup> year<sup>-1</sup> (Ellermann et al., 2015).



**Fig. 7.1.2.2-1.** Cumulative probability distribution of Cd in topsoil and bottom soil from the Baltic Soil Survey (Reimann et al., 2003). Please note the log scale on the x-axis and the probit scale on the y-axis.

#### 7.1.3 Predicting Cd input to soil

For predicting Cd input to soil, given as g Cd ha<sup>-1</sup> yr<sup>-1</sup>, in the four selected regions, the following information was used: information from the local agricultural extension service about common crop rotations and common application of P fertilisers and manure to the different crops; data from NFSA on sale of lime products in the counties where the case areas are located; data from SSB on application of sewage sludge in the case areas;

measured concentrations of Cd in fertilisers commonly used in Norway (Table 7.1.2.1.1-1); average Cd content of manure (Table 7.1.2.1.2-1) and lime products (section 7.1.2.1.4); average Cd concentration in sewage sludge (Section 7.1.2.1.3); maximum allowed application of sewage sludge (FOR 2003-07-04 nr 951), and predicted Cd atmospheric contribution (Eq. 7).

**Cd input via Sewage sludge:** As the practice of sewage sludge application has a strong influence on the total Cd contribution to soil, we chose to calculate with both the use of maximum allowed sewage sludge application according to the regulation (40 tonnes ha<sup>-1</sup> 10 yr<sup>-1</sup> for sewage sludge concentration of 0.6 mg Cd kg<sup>-1</sup> DW), which might be the case for some fields (represent a maximal conservative and realistic worst case scenario), and the average application of sewage sludge for the total agricultural area in the case municipalities, based on information from SSB. Using the maximal dose according to the regulation, the annual input of Cd is predicted to be about 2.4 g Cd ha<sup>-1</sup>. With use of average application of the sewage sludge, the predicted annual input is 0.033 mg Cd ha<sup>-1</sup> and 0.021 mg Cd ha<sup>-1</sup> at Ås and Stange (cultivation of cereals), respectively.

**Cd input via mineral P fertiliser:** The predicted Cd contribution to agricultural soil via mineral P fertilisers under today's agricultural practices, and most used NPK in these cases (range 25-50 mg Cd kg<sup>-1</sup> P), is 0.14-0.17 g Cd ha<sup>-1</sup> yr<sup>-1</sup> for production of grass, 0.32-0.42 g Cd ha<sup>-1</sup> yr<sup>-1</sup> for production of cereals, and 0.99 g Cd ha<sup>-1</sup> yr<sup>-1</sup> at Stange with a crop rotation with potato, carrot, and cereals (Table 7.1.3-1).

With today's agricultural practice, an increase in the Cd concentration in mineral fertiliser to a maximum limit of 45.8, 91.6, and 137.4 mg Cd kg<sup>-1</sup> mineral P fertiliser, results in a predicted increase in Cd contribution from mineral fertiliser in the range of 0.07 to 0.35, 0.32-1.24, and 0.55-2.35 g Cd ha<sup>-1</sup> yr<sup>-1</sup>, respectively.

At Ås, Stange, Time and Melhus, all with cultivation of cereals, ML at 137.4 mg Cd kg<sup>-1</sup> mineral P fertiliser would result in a predicted 5 to 8 times increase in Cd input compared to today's fertilising practice and Cd content in Norwegian mineral P fertilisers (NFSA, 2017).

Even if the reported application of mineral P fertilisers in EU used in the risk evaluation by Six and Smolders (2014) were lower than in this risk assessment, the Cd concentration in fertilisers used in the EU risk evaluation was higher, 36 mg Cd kg<sup>-1</sup>  $P_2O_5$  which corresponds to 82.4 mg Cd kg<sup>-1</sup> P (from Fertilisers Europe, 2011 in Six and Smolders, 2014). This results in a predicted higher Cd input to agricultural soil in the risk evaluation by Six and Smolders (Six and Smolders, 2014) 0.8 g Cd ha<sup>-1</sup> for cereal production and 1.6 g ha<sup>-1</sup> for potato, compared to the predicted input in the four case municipalities in Norway. In a study from 2008 (Nziguheba and Smolders, 2008), n=196 samples including both pure and blended samples), the Cd concentration in P mineral fertilisers range from < 0.1 to 120 mg Cd kg<sup>-1</sup>  $P_2O_5$ , corresponding to 0.2 – 254.8 mg Cd kg<sup>-1</sup> P.



**Table 7.1.3-1.** Amount of Cd added to agricultural soil, given in g Cd ha<sup>-1</sup> yr<sup>-1</sup>.

Munic	cipalities	Ås¹	Stange <sup>1</sup>	Stange <sup>2</sup>	Time <sup>3</sup>	Melhus <sup>1</sup>	Melhus <sup>3</sup>	
		Cd contribution via other sources (g Cd ha <sup>-1</sup> yr <sup>-1</sup> )						
Atmo	spheric	0.34	0.34	0.34	0.34	0.34	0.34	
Lime		0.01	0.01	0.01	0.02	0.01	0.01	
Sewa sludg		2.40	2.40	6)				
Sewa sludg		0.03	0.02	6)				
Manu	re			0.54	0.24	0.37		
		Cd cd	ontribution	mineral P	fertilisers	(g Cd ha <sup>-1</sup>	yr <sup>-1</sup> )	
8	25-50 <sup>8</sup>	0.37	0.42	0.99	0.14	0.32	0.17	
ng (	45.8	0.68	0.76	1.11	0.23	0.58	0.24	
ML <sup>7</sup> (mg ( kg <sup>-1</sup> P)	91.6	1.36	1.53	2.23	0.46	1.15	0.49	
Σ	137.4	2.04	2.29	3.34	0.69	1.73	0.73	
		Sum C	d input wi	th maximu	ım allowed	l sewage s	ludge <sup>4</sup>	
8	25-50 <sup>8</sup>	3.12	3.16	1.34	1.04	0.91	0.90	
ng (	45.8	3.43	3.51	1.46	1.13	1.17	0.97	
ML <sup>7</sup> (mg (kg <sup>-1</sup> P)	91.6	4.11	4.27	2.57	1.36	1.75	1.21	
Σ	137.4	4.79	5.04	3.69	1.59	2.32	1.46	
		Sum Cd input with average applied sewage sludge <sup>5</sup>						
8	25-50 <sup>8</sup>	0.75	0.78	1.34	1.04	0.91	0.90	
ng (	45.8	1.06	1.13	1.46	1.13	1.17	0.97	
ML <sup>7</sup> (mg ( kg <sup>-1</sup> P)	91.6	1.74	1.89	2.57	1.36	1.75	1.21	
Σ	137.4	2.42	2.66	3.69	1.59	2.32	1.46	

<sup>&</sup>lt;sup>1</sup> Crop rotation cereals, <sup>2</sup> Crop rotation potatoes, carrots, cereals, <sup>3</sup> Grass, <sup>4</sup> Predicted based on use of maximum allowed sewage sludge applied according to regulation for sewage sludge quality class I, 40 tonnes DW ha<sup>-1</sup>10yr<sup>-1</sup> (Table 3.1.1-1); <sup>5)</sup> Predicted based on average application of sewage sludge, data from 2008-2017 (SSB); <sup>6)</sup> Sewage sludge is not included, because according to current regulation, application is not allowed in crop rotations with less than three years between application and growing potato/vegetables (FOR 2003-07-04 nr 951), <sup>6)</sup> Sewage sludge is not included, because according to current regulation, application is not allowed in crop rotations with less than three years between application and growing potato/vegetables (FOR 2003-07-04 nr 951), <sup>7</sup> Maximum level values for Cd concentration in mineral P fertilisers (mg Cd kg<sup>-1</sup> P), <sup>8</sup> Measured Cd concentration in Norwegian fertilisers (NPK 25:2:6, 22:3:10, 12:4:8) used in this risk assessment (range 2-50 mg Cd kg<sup>-1</sup> P).

#### 7.1.4 Loss of Cd from soil via leaching and plant harvesting

#### Kinetic rates

The kinetic rates of loss of Cd from soil, including loss via leaching (predicted using Eq. 2) and removal via harvesting of plants (predicted using Eq. 5), are presented in Table 7.1.4-1.

These kinetic rates are further used for predicting loss of g Cd ha<sup>-1</sup> yr<sup>-1</sup> (Table 7.1.4-2). The parameters used for predicting leaching rate (k), Kd and  $K_{\text{soil-water}}$ , soil organic matter (SOM) and OC, are shown in Table 8.1-1.

The predicted total removal rate of Cd from soil,  $\sum k_{leaching+}k_{plant-removal}$ , was highest in Time, at 0.058 yr<sup>-1</sup> (grass production), followed by Melhus, at 0.014 yr<sup>-1</sup> (grass and cereal production). The predicted exceptionally high removal rate at Time is connected with high annual precipitation and precipitation excess. In addition, mean soil organic matter (SOM) is 4.1% (corresponding 2.4 % OC) and mean pH 5.3 (lowest of the four regions, Table 8.1-1), thereby affecting the Kd value, which was 115.0 L kg<sup>-1</sup> compared to Kd 217.6-257.8 in the other case regions.

In Ås, the predicted total loss rate was 0.0052 yr<sup>-1</sup>, and in Stange, 0.0046 yr<sup>-1</sup> and 0.0050 yr<sup>-1</sup> for crop rotations of solely cereals and combinations of cereals, potato, and carrot, respectively.

**Table 7.1.4-1.** Removal kinetic for loss via leaching and crop harvesting for each of the municipalities, given as  $yr^{-1}$  ( $k_{leaching} = k_l$ ,  $k_{plant-removal} = k_p$ ).

Removal rate yr <sup>-1</sup>	Ås¹	Stange <sup>1</sup>	Stange <sup>2</sup>	Time <sup>3</sup>	Melhus <sup>1</sup>	Melhus <sup>3</sup>
Leaching, k <sub>l</sub>	0.0049	0.0043	0.0043	0.0545	0.0140	0.0140
Plant harvest, kp	0.0003	0.0003	0.0007	0.0033	0.0002	0.0009
∑kı+k <sub>p</sub>	0.0052	0.0046	0.0050	0.0579	0.0142	0.0149

<sup>&</sup>lt;sup>1</sup> Crop rotation cereals, <sup>2</sup> Crop rotation potatoes, carrots, cereals, <sup>3</sup> Grass.

In spite of the highest predicted Kd value at Melhus (Table 8.1-1), the removal rate is high due to high precipitation and infiltration rate (precipitation excess) as compared to Ås and Stange.

At all case sites, the loss rate via leaching was higher than via plant uptake, by around 3 to 64 times. The least difference between leaching and harvest was predicted for crop rotation with potatoes and carrot in Stange. The highest difference was predicted in Melhus with crop rotation with cereals. This due to generally to lower removal via cereals and a rather high leaching rate due to high precipitation and precipitation excess.

#### Predicted loss of Cd per ha per year

In the risk evaluation by Smolders (2013), pore water concentrations of Cd (predicted from soil concentration and Kd) and precipitation excess were used for estimating loss of Cd per ha per year (Eq. 4). In order to compare this approach with leaching calculated with equation 2, this was also performed.

Loss of Cd via leaching predicted via equation 4 (section 6.2.2) ranged from 3.5 to 3.7 g Cd ha<sup>-1</sup> yr<sup>-1</sup> in Ås and Melhus, but as high as 18.5 and 19.4 g Cd ha<sup>-1</sup> yr<sup>-1</sup> in Stange and Time. The exceptionally high predicted Cd loss in Stange is due to the high background

concentration of 1.7 mg Cd kg<sup>-1</sup> DW. The very high loss of Cd in Time is, as discussed above, is related to very high precipitation and precipitation excess, and predicted low Kd.

The enhanced leaching of Cd due to an increased maximum level of Cd in mineral P fertilisers was predicted to be lower than the predicted leaching of Cd from background soil; from 0.04% in Stange, with a high background Cd concentration in soil, to 0.2-0.5% in the other case sites.

**Table 7.1.4-2.** Removal of Cd via leaching and harvesting of plants, presented separately for removal of Cd in current background soil and Cd added via mineral P fertilisers, at different regions. Note different units for Cd removal from background and mineral fertilisers.

	Removal	Ås²	Stange <sup>2</sup>	Stange <sup>3</sup>	Time <sup>4</sup>	Melhus <sup>2</sup>	Melhus <sup>4</sup>
	Leaching <sup>1</sup>	3.5	18.5	18.5	19.8	3.6	3.6
Cd in back-ground soil (g Cd ha <sup>-1</sup> yr <sup>-1</sup> )	Plant harvest	0.24	1.26	2.90	1.21	0.06	0.23
	Σleaching <sup>1</sup> + harvesting	6.0	32.0	33.7	34.1	6.1	6.3
	Leaching <sup>1</sup>	3.0	3.0	7.1	12.3	4.0	4.0
Current min. P practice (mg Cd ha <sup>-1</sup> yr <sup>-1</sup> )	Plant harvest	0.1	0.1	0.7	0.5	0.04	0.2
	Σleaching <sup>1</sup> + harvesting	3.2	3.1	7.8	12.7	4.1	4.2
	Leaching <sup>1</sup>	16.6	16.4	24.0	62.4	53.7	17.1
Highest ML in min. P <sup>5</sup> (mg Cd ha <sup>-1</sup> yr <sup>-1</sup> )	Plant harvest	0.7	0.7	2.3	2.3	0.5	0.7
	Σleaching+ harvesting	17.3	17.1	26.3	64.6	54.3	17.7-1

<sup>&</sup>lt;sup>1</sup> Predicted with Eq. 2, <sup>2</sup> Crop rotation cereals, <sup>3</sup> Crop rotation potatoes, carrots, cereals, <sup>4</sup> Grass <sup>5</sup> ML 137.4 mg Cd kg<sup>-1</sup> P.

## 7.2 Transfer to crops and animal feed

In order for Cd to enter the food chain, the Cd must accumulate in the portion of the plant that is consumed. Plant uptake of Cd is variable and depends on a number of parameters, among them the soil properties (especially pH, soil organic matter, SOM, and texture, and thus  $K_d$  value) and properties of the plant itself (species, plant parts, age etc.) (e.g., McLaughlin et al., 2011; Yang et al. 1995; He and Singh 1994).

Regarding differences between plant species, the concentration of Cd in different plant species when grown in the same soil varied in the order of spinach>carrot>rye grass>oat (Kashem and Singh, 2002). For example, Cd concentrations in oat, carrot, and spinach were 74, 115, and 1155 µg kg<sup>-1</sup>, respectively. Similarly, carrot leaves contained 5-6 times more Cd than carrot roots, and oat straw contained 2-3 times as much Cd as oat grain (He and Singh 1994). Genetic variation in Cd uptake has been reported in carrot (Harrison 1986), potato (Sparrow et al. 1992), wheat (Oliver et al. 1995), and cucumber (Harrison and Staub 1986). Among durum wheat varieties in Canada, Cd in grain averaged 0.77 mg Cd kg<sup>-1</sup> for Arcola

variety, 0.157 mg Cd kg<sup>-1</sup> for Kyle variety, 0.038 mg Cd kg<sup>-1</sup> for Genesis variety, and 0.055 mg Cd kg<sup>-1</sup> for Katepwa variety (Grant et al. 1998). In a report from Food Standards Agency it is reported a higher Cd concentration in baked potato skin compared to baked potato flesh (FSA, 2012).

It is also found that although moraine alum shale (loam) soil contained lower total Cd (1.76 mg kg $^{-1}$ ) than alum shale clay loam (2.80 mg kg $^{-1}$ ), Cd concentrations in wheat grain grown in alum shale soils, was generally the same (0.42 µg kg $^{-1}$ ) (Singh et al., 1995). This implies that Cd transfer factor was much higher in moraine (0.24) than in clay loam (0.15). This further indicates that soil type, and especially soil texture, plays an important role in transferring metals from soil to plants.

All the different parameters influencing uptake in plants are rarely controlled or measured, which is why the simpler approach of constant TF was chosen. Moreover, multivariate regressions considering soil pH, organic matter, and clay content were not superior to simple TF when tested with a large dataset (Francois et al. 2009; He and Singh, 1993; Legind and Trapp, 2010). Empirically derived TF (µg Cd per kg fresh weight plant/µg Cd per kg soil) were also used by Smolders (2013) in the risk assessment on Cd in mineral fertilisers. As discussed by Six and Smolders (2014), as even the increase of Cd uptake in crops with increasing Cd concentration in soil is less than proportional (Eriksson et al., 1996), it is justifiable to assume proportionality of Cd concentrations in crops and soil.

For this risk assessment, different methods for estimating plant uptake were evaluated. These included: TFs used by Smolders (2013), regression to soil pH and soil Cd concentration (ERM, 2000), and a mechanistic model (Legind et al., 2012) with Kd by Smolders (2013). These methods and their comparison are described in Appendix IV. The comparison was made for six Norwegian regions using data from the risk assessment of Amundsen et al. (2000). Based on this evaluation, TFs from Legind et al. (2012) for leafy vegetables and grass, and from Smolders (2013) for carrot, potato, and cereals were selected for this risk assessment.

The concentrations of heavy metals in crops are typically measured in DW, whereas human consumption data are given in FW. Plant-specific water content (WC) are given in Appendix IV. The values selected for this risk assessment are shown in Table 7.2-1.

For predicting Cd exposure of farm animals and humans, the Cd concentrations in FW food and DW forage are used.

**Table 7.2-1.** Transfer factors (TF) for Cd, selected and applied for estimating plant concentrations and water content (WC) used in this risk assessment is shown. TF is given as mg Cd kg<sup>-1</sup> DW plant tissue over mg Cd kg<sup>-1</sup> DW soil and WC in %. References and % WC used by the different authors are also presented.

Plant	TF (mg Cd kg <sup>-1</sup> DW plant tissue over mg Cd kg <sup>-1</sup> DW soil)	WC (%)	References
Leafy vegetables	0.36	70	US EPA, cited in Legind and Trapp (2010)
Carrot	0.25	70	Legind and Trapp (2010), WC 88%
Potatoes	0.27	70	Smolders (2013), WC 77.8%
Cereals	0.165	15	Smolders (2013), WC 15%
Grass	0.36	80	US EPA, cited in Legind and Trapp (2010)

## 7.3 Toxicity of cadmium

Cadmium is toxic to humans and various forms of animal life and is generally not considered an essential nutrient. It has specific adverse effects on the kidney, and may induce hypertension and microcytic, hypochromic anaemia. However, in studies on rodents, chickens, and livestock, addition of low levels of Cd to their diets have been shown to increase weight gain (Bokori and Fekete, 1995). The basis of these effects is mainly unknown, but may result from antibiotic or pharmacological actions as observed with other trace elements, such as Zn and Cu. Furthermore, Cd is an essential nutrient for the common marine diatom, *Thalassiosira weissflogii*, where Cd is a cofactor for an isoform of carbonic anhydrase. The enzyme is needed under conditions of low Zn – which is often the case in many marine environments (Lane and Morel, 2000). Higher plants have not been shown to need Cd (NRC, 2005).

The toxicity of Cd is partly alleviated by high dietary Zn, Fe, and Ca, probably via a complex interaction between these three elements in Cd protection. The toxic effects of Cd are thought to be caused by free Cd ions. Cadmium bound to metallothionein is usually less active (NRC, 2005).

### 7.3.1 Predicted no-effect concentrations (PNEC) in the environment

In order to assess the risk of adverse environmental effects posed by application of fertilisers containing Cd, the maximum level of Cd that is not expected to cause a toxic effects needs to be established for the environmental compartments exposed, e.g., soil and surface water. These concentration levels are referred to as PNECs.

PNECs for Cd in surface water and soil have recently been proposed in a risk assessment report (RAR) within the framework of Council Regulation 793/93/EEC on existing chemicals (EC, 2007). The PNECs are based on compiled data on toxicity of Cd to various aquatic and terrestrial organisms according to criteria and principles described in a technical guidance documents (ECB, 2003; ECHA, 2016). The same database has been used to establish environmental quality standards (EQS) under the water Framework Directive. In the present

risk assessment of Cd in Norwegian agricultural soils and surface waters, PNECs from the European RAR are used. A brief summary of the basis and procedures used to derive the PNECs for the aquatic and terrestrial environments is presented in the following paragraphs.

#### 7.3.1.1 Terrestrial

The data selected for derivation of PNEC for soil organisms that are exposed to Cd in soil include NOECs from long-term tests with microorganisms, plants, and soil fauna exposed to Cd in soil. The tests with microorganisms are performed on the native microflora in soil, where effects of Cd on important element-cycling processes (e.g., respiration, ammonification, or nitrification) or enzyme activity are studied. For plants, the endpoints are seed germination or growth of roots or shoots. Various earthworms and springtails are used as representatives of the soil fauna in tests in which the effects on growth or reproduction are investigated.

For microorganisms, 36 tests were selected as a basis for PNEC derivation. The NOECs spanned from 3.6 mg Cd kg<sup>-1</sup> DW (soil dry weight) to 3000 mg Cd kg<sup>-1</sup> DW.

Among 37 selected tests on soil fauna, the NOECs ranged from 5 mg Cd kg<sup>-1</sup> (cocoon production in *Eisina fetida*) to 320 mg Cd kg<sup>-1</sup> (weight gain in *Folsomia candida*).

For plants, 54 tests were selected. The lowest NOEC (1.8 mg Cd kg<sup>-1</sup>) was found in a test where effects on the root length of *Picea sitchensis* was studied. The highest NOEC in tests with higher plants was 160 mg Cd kg<sup>-1</sup>. The lower range of effect concentrations for plants as compared with microbial processes and soil fauna may indicate that higher plants are generally the most sensitive group of soil organisms exposed to Cd in soil.

#### 7.3.1.2 PNEC<sub>soil</sub> based on direct exposure to soil

In the Cd RAR, the NOECs from all selected tests with microorganisms, plants, and soil fauna were pooled for an analysis of species-sensitivity distribution. The calculated HC5 was 2.3 mg Cd kg<sup>-1</sup>, None of the tests included showed toxic effects at concentrations as low as 2.3 mg Cd kg<sup>-1</sup>. The lowest LOEC was 2.5 mg Cd kg<sup>-1</sup> in tests on several plant species. The RAR did not come to a firm conclusion regarding an AF for calculation of PNEC<sub>soil</sub> from the HC5, and the recommendation was to apply an AF between 1 and 2, which yields PNEC<sub>soil</sub> of 1.15-2.3 mg Cd kg<sup>-1</sup> soil DW.

The toxicity data on which the PNEC<sub>soil</sub> is based stem from tests where various terrestrial organisms have been exposed to soil spiked with Cd in the form of CdO or  $Cd^{2+}$  salts. The test organisms were introduced to the spiked soils after a relatively short time. There is, however, strong evidence that the biological availability of metals in soil is often reduced with time due to stronger binding to the soil matrix. This phenomenon, which may involve different processes, is known as ageing and is discussed in greater detail in section 6.2.3. Because of the effect of ageing, PNEC<sub>soil</sub>, which is based on toxicity tests in freshly spiked soils, is likely to be overprotective when used in risk assessment of metals that have

accumulated in soil over a long period (Lock and Janssen, 2003). In the present risk assessment, the accumulation of Cd in soil after up to 100 years after application of fertilisers is calculated. In this perspective, ageing is likely to be a significant factor influencing the bioavailable fraction of Cd in soil. Hence, the least conservative option for AF (1) recommended in the RAR was used for calculation PNEC from the HC5:

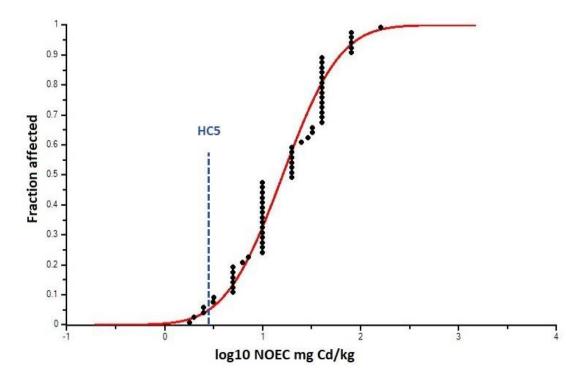
 $PNEC_{soil} = HC5/1 = 2.3 \text{ mg Cd kg}^{-1} \text{ soil, dry weight.}$ 

## 7.3.1.3 PNECterrestrial plants

No separate PNEC for plants was calculated in the RAR, where the effect concentrations from toxicity tests with plants were pooled with those from microorganisms and soil fauna for deriving a PNEC<sub>soil</sub>.

In order to show whether the  $PNEC_{soil}$  is sufficiently protective for plants, a separate analysis of the sensitivity distribution of the NOECs from tests with plants was conducted here, as agricultural plants are one of the specific targets for risk assessment according to the Terms of reference for this opinion.

The database includes 41 plant NOEC values that were used for derivation of PNEC $_{soil}$ . The RAR notes that few studies were found in recent literature. A literature search was performed to see whether additional studies have been published since the RAR database was compiled. This resulted in 19 additional NOECs for eleven species of terrestrial plants (da Rosa Corrêa et al. 2006, de Oliveira et al. 2016). Three of these species were not included in the RAR (See Table A-V-1 Appendix V). An SEM was applied to the NOEC data, calculating the median 5th percentile (HC5) of the log-normal distribution with the software package ETX 2.2 (RIVM, Bilthoven, The Netherland). The HC5 was calculated at 2.79 mg Cd kg<sup>-1</sup> (See Fig. 7.3.1.3-1).



**Fig. 7.3.1.3-1.** The cumulative frequency distribution of the selected NOEC values of Cd toxicity tests of terrestrial higher plants. Observed data and logistic distribution curve for the whole data set fitted on the data.

The tests on which the HC5 value is based were performed in soils with pH 3.3-7.9, % carbon 0.6-45, and % clay 3-41. There is, however no significant correlation between these parameters and the NOECs for plants, although the two lowest NOECs were found in soils with the lowest pH (pH 3.2 and 3.9 respectively).

The HC5 is based on 60 NOECs representing 16 species (See Table A-V-1 Appendix V). In these tests, only one LOEC was lower than the HC5. This test was performed on Sitka spruce (*Picea sitchensis*) in an acid forest soil at pH 3.2. Clearly, such soil should not be compared with arable soils at higher pH values for which higher LOECs were found. In order to allow comparison with the generic PNEC<sub>soil</sub> for terrestrial organisms, the same AF (1) is proposed for derivation of PNEC<sub>soil</sub>, plants. Hence:

PNEC<sub>soil, plants</sub> =  $HC5/1 = 2.8 \text{ mg Cd kg}^{-1} \text{ soil, dry weight.}$ 

#### 7.3.1.4 PNEC<sub>soil</sub> in relation to soil properties

No empirical equation has been developed for Cd to normalize toxicity to a standard soil. It is often assumed that the metal concentration in soil solution represents the toxic dose for the ecosystem and, therefore, a correlation between metal toxicity and pH is to be expected. At higher pH, metal solubility is low and the HC5 could be higher than at low pH where metals are more soluble. A significant correlation between the log NOEC values and soil pH was, however, not found in the data collected for the RAR. A positive trend between log NOEC and soil pH emerges up to about pH 6, beyond which there seems to be no further trend. For organic carbon, which is also known to affect the mobility of metals in soil, the data were insufficient to perform an analysis of its effect on NOEC values in the database.

The tests on which the HC5 value is based were performed in soils with pH 3.1-7.9, % carbon 0.6-47, and % clay 2-70. This range of soil properties covers most of the European topsoils, and the PNEC<sub>soil</sub> of 2.3 mg Cd kg<sup>-1</sup> has been proposed as a generic PNEC for soils in Europe.

### 7.3.1.5 PNEC<sub>soil</sub> based on secondary poisoning in mammals

Studies on soil-dwelling animals have shown that accumulation of Cd occurs from the soil, resulting in higher concentrations in the organisms than in the surrounding soil. Earthworms, which ingest soil, are particularly exposed to Cd via this route, and bioaccumulation factors up to 150 (median 15) have been reported for earthworms according to data reviewed in the RAR. This shows that secondary poisoning within the terrestrial food chain should also be considered in establishment of PNEC<sub>soil</sub>.

In the RAR, kidney Cd concentrations of wildlife as an indicator of Cd exposure and risk were used to derive a PNEC<sub>soil</sub> that accounts for secondary poisoning. The kidney is regarded as the critical organ in chronic Cd toxicity. With continued exposure, there is a continual increase in Cd concentrations in the renal cortex until a critical value is reached and histopathological changes and renal dysfunction occur (proximal tubular cell necrosis, proteinuria, and glycosuria; Scheuhammer (1987) and references therein). This critical value should be regarded as a sub-lethal endpoint. The risk of secondary poisoning can be assessed by calculating the exposure at which this critical value is not exceeded for wildlife.

WHO has suggested 100-200  $\mu$ g Cd g<sup>-1</sup> wet weight in the renal cortex as a critical range and the lowest limit of this range was used in the assessment in the RAR, i.e., 100  $\mu$ g Cd g<sup>-1</sup> wet weight in the renal cortex, which corresponds with a whole kidney, DW-based value of 400  $\mu$ g Cd g<sup>-1</sup> DW.

Kidney-Cd concentrations in wildlife were compiled in the RAR. The data include mean or median Cd concentrations for 9 different mammalian species (no data found for birds). Paired sets of soil Cd/kidney-Cd were found for 8 mammal species from areas differing in soil Cd concentrations and pH. Most of these studies have measured the Cd concentrations at two or more locations with differing metal exposures. The critical soil Cd concentration is

defined as that concentration at which a critical kidney Cd concentration of 400  $\mu$ g Cd g<sup>-1</sup> DW (whole kidney) is predicted, using a proportional extrapolation from each paired soil/kidney Cd concentration set. The lowest critical soil Cd concentration was selected from each transect study, or when only a range in soil or kidney Cd concentrations was available. The lowest critical concentrations were found in moles and shrews, which feed on earthworms and insects. Herbivore species, such as voles and rabbits, showed higher critical concentrations. Furthermore, the data showed that the critical concentrations were lowest in acidic soils. The lowest critical concentration in the dataset was 0.6  $\mu$ g Cd g<sup>-1</sup> DW in a mole from an area with low soil pH.

The cumulative frequency of the critical soil Cd concentration, at which the critical kidney Cd concentration (400  $\mu$ g Cd g<sup>-1</sup> DW) may be exceeded in the average population of different species, was analysed and the log-logistic curve fitted on all individual data (n=20). The statistical extrapolation method predicts that the HC5 for protecting mammals is 0.9  $\mu$ g Cd g<sup>-1</sup> DW.

The HC5 for protecting mammals is about twofold below the PNEC<sub>soil</sub> derived for protecting plants, soil fauna, and microflora. This assessment therefore suggests that biotransfer of Cd from soil to higher trophic levels is the most critical pathway for Cd.

PNEC<sub>soil, secondary poisoning</sub> = HC5<sub>critical soil concentration</sub> = 0.9 mg Cd kg<sup>-1</sup> soil, dry weight

#### 7.3.1.6 Aquatic

A wealth of data exists on the toxic effects of Cd to freshwater organisms. Only studies presenting no observed effect concentrations (NOEC) for chronic toxicity were included as a basis for PNEC. For species where many NOECs were available, geomean values were calculated for NOECs representing the same endpoint and test medium.

#### 7.3.1.7 Fish and amphibians

Nineteen tests of chronic effects of Cd on fish and amphibians were selected as a basis for derivation of aquatic PNECs. In general, toxicity is most pronounced in soft water. Reproduction parameters are most sensitive to Cd. The lowest reported chronic effect concentration for fish was  $0.8~\mu g~L^{-1}$  in a test of reproduction of Atlantic salmon (*Salmo salar*).

#### 7.3.1.8 Aquatic invertebrates

Twenty-two tests of chronic toxicity in invertebrates from which NOEC could be derived were selected. Certain Cladocera (e.g., *Daphnia* and *Ceriodaphnia*), appear to be particularly sensitive to Cd. Most chronic lowest-observed effect concentration (LOEC) values of *Daphnia* range between 1 and 10 µg L<sup>-1</sup>. The lowest LOEC value was found in a reproduction test with

Daphnia magna, where the mean number of young per adult after 21 days of exposure was reduced at 0.29  $\mu$ g L<sup>-1</sup> (LOEC). The NOEC in this test was 0.16  $\mu$ g L<sup>-1</sup>.

#### 7.3.1.9 Algae and aquatic plants

Eight tests with 6 species of plankton algae and one plant ( $Lemna\ paicicostata$ ), were selected. The data show that Cd can affect primary producers in the 1-10  $\mu$ g L<sup>-1</sup> range, but no tests showed toxicity below 1  $\mu$ g L<sup>-1</sup>. Algae are likely to be most sensitive to Cd at nutrient-limiting conditions and low cell density-. With one exception, all tests were performed in artificial media, some of which had a very similar composition as freshwater samples. The lowest LOEC (1.9  $\mu$ g L<sup>-1</sup>) was found for growth of the diatom *Asterionella formosa* (NOEC 0.85  $\mu$ g L<sup>-1</sup>).

#### 7.3.1.10 **PNEC**<sub>water</sub>

The RAR considered that the diversity of the data (NOEC values from 28 species and 16 different families, including warm and coldwater fish, amphibians, crustaceans, algae, and higher plants) was large enough to use the statistical extrapolation method (SEM) to calculate the PNEC. Thus, the PNEC<sub>water</sub> for Cd was derived from the median 5<sup>th</sup> percentile (HC5) from the distribution of 44 chronic NOEC values, some of which are geometric species means. The NOEC values were obtained from laboratory-based, single-species studies and refer to the dissolved fraction of Cd.

The SEM (Aldenberg and Slob, 1993) was applied on the NOEC data, and HC5 estimated from the log-logistic and the log-normal distributions Both distribution models showed a HC5 at 0.38 µg Cd L<sup>-1</sup>. A generic PNEC was derived from the HC5, including an assessment factor (AF) of 2 to account for remaining uncertainty, yielding:

#### Generic PNEC<sub>water</sub> = $HC5/2 = 0.19 \mu g L^{-1}$

None of the 168 tests underlying the derived PNEC showed a toxic effect as low as 0.19  $\mu$ g Cd L<sup>-1</sup>. The lowest LOEC, found in a test on *Daphnia magna*, was 0.28  $\mu$ g/L.

#### 7.3.1.11 PNEC<sub>water</sub> accounting for water characteristics

The toxicity of metals in water is often modified by abiotic factors, such as pH, dissolved organic carbon (DOC), and water hardness. For some metals Biotic Ligand Models (BLM) have been developed to describe the effects of abiotic factors on toxicity (Paquin et al., 2002). By using BLM-models, NOECs from tests performed in different media can be normalised to a specific combination of abiotic factors, in order to derive region-specific PNECs. For Cd, no complete BLM model is currently available, but sufficient data are available to describe the effect of hardness on chronic toxicity to aquatic organisms. The generic PNEC $_{water}$  is based on toxicity tests in various aquatic media, which differ with respect to pH, hardness, and organic carbon content. Hence, the generic PNEC $_{water}$  (0.19  $\mu$ g L $^{-1}$ )

might not be protective for waters with very low hardness. The RAR therefore suggests using specific, refined PNECs for regional risk assessments.

A water hardness correction equation of the United States Environmental Protection Agency (US EPA) (US EPA, 2001) was used to calculate the HC5 as a function of water hardness (Table 7.3.1.11-1). All NOEC values were converted to NOEC values at a reference hardness of 50 mg  $CaCO_3$  L<sup>-1</sup>, and the HC5 recalculated. Based on the hardness-normalized HC5, and using an AF of 2, the PNEC<sub>water 50H</sub> was calculated to be 0.09  $\mu$ g Cd L<sup>-1</sup>. Using the hardness-correction equation, regional PNECs for different hardness levels were calculated from the HC5 with an AF of 2 as shown in Table 7.3.1.11-1.

**Table 7.3.1.11-1.** The influence of hardness (mg CaCO<sub>3</sub> L<sup>-1</sup>) at PNEC (EC, 2007).

Hardness (mg CaCO <sub>3</sub> L <sup>-1</sup> )	PNEC (µg Cd L <sup>-1</sup> )
40	0.08
50	0.09
100	0.15
200	0.25

In an addendum to the European Union RAR, the issue of risk characterisation of Cd in very soft waters (hardness below 40 mg  $CaCO_3$  L<sup>-1</sup>) has been addressed. The report concluded that no further adjustment of the PNEC in very soft water is necessary and the previously agreed PNEC of 0.08  $\mu$ g L<sup>-1</sup> is proposed for waters with hardness 2.7-40 mg  $CaCO_3$  L<sup>-1</sup> and dissolved organic carbon concentrations above 2 mg C L<sup>-1</sup> (EC, 2008). The equation used for hardness normalization should not be extrapolated to water hardness below 40 mg  $CaCO_3$  L<sup>-1</sup>.

In Norwegian surface waters, the hardness level is generally low. A survey of 1006, mostly pristine, lakes in Norway showed a medium value of only 4 mg CaCO<sub>3</sub> L<sup>-1</sup> and 90% of the lakes had a hardness less than 18 mg CaCO<sub>3</sub> L<sup>-1</sup> (Skjelkvåle et al., 2001). Surface waters in agricultural areas are likely to show slightly higher hardness levels, but still, in most cases, <50 mg CaCO<sub>3</sub> L<sup>-1</sup>. Thus, a **PNEC**<sub>water</sub> = **0.08**  $\mu$ g **Cd** L<sup>-1</sup> will be used for risk assessment in Norwegian surface waters.

#### 7.3.1.12 PNEC<sub>sediment</sub>

Sediment-dwelling organisms may be exposed to Cd in the sediment via uptake across respiratory surfaces and body walls from the porewater, or by ingestion of sediment. In aerobic sediments, the availability of Cd for biological uptake is mainly affected by the content of organic carbon and Fe and Mn-hydroxides. In anaerobic sediments, acid-volatile sulphides reduce bioavailability and toxicity by binding and immobilising Cd as insoluble sulphide.

Only limited relevant data on the toxicity of Cd to freshwater benthic organisms were found in the review performed for the RAR (ECB, 2007). Seventeen tests were selected as a basis

of PNEC estimation. These data refer to tests in which uncontaminated sediment was spiked with Cd²+ salts. Only one of the sediment toxicity tests available within the dataset can be considered as a real chronic test. This is a test in freshwater sediments spiked with Cd, which showed a NOEC of 115 g kg⁻¹ dry weight on the abundance of chironomids (*Chironomus salinarius*) after 14 months (Hare et al., 1994). This NOEC was used as a basis for PNEC calculation, with an AF of 50. The choice of an AF of 50 instead of 100 is justified by the number of acute toxicity data, showing little differences between species. This results in:

## $PNEC_{sediment} = 115 \text{ mg kg}^{-1}/50 = 2.3 \text{ mg Cd kg}^{-1}DW$

According to the TGD (ECB, 2003) PNEC<sub>sediment</sub> may also be calculated using the equilibrium partitioning method in the absence of ecotoxicological data for sediment-dwelling organisms. The formula used for the calculation is:

 $PNEC_{sediment} = K_P \cdot PNEC_{water} \cdot 10^{-3} \text{ mg/kg DW}$ 

Where Kp equals the solid/water partitioning coefficient of suspended matter expressed in L kg<sup>-1</sup> and PNEC<sub>water</sub> is expressed in  $\mu$ g L<sup>-1</sup>. For chemicals with K<sub>P</sub> > 2000, which is the case for Cd, an additional safety factor of 10 should be included to take the risk of direct ingestion into account. Using a typical Kp value of 130000 for Cd (EC 2007) and the generic PNEC<sub>water</sub> of 0.19  $\mu$ g L<sup>-1</sup>, gives PNEC<sub>sediment</sub> = 2.5 mg Cd kg<sup>-1</sup> DW. This estimate supports the PNEC<sub>sediment</sub> derived from ecotoxicological data for sediment-dwelling organisms as shown above.

#### 7.3.2 Cd toxicity in farm animals

The toxicity of Cd is well recognised, and even low exposure accumulates in the body. Primarily in order to prevent high levels in animal food products, WHO has set a 1 mg/kg upper limit in complete feeds for animals (IPCS, 1992a).

The bioavailability of Cd depends on its chemical form. Cadmium in ionic form and as salt, such as cadmium chloride, have highest bioavailability, but in general Cd is not absorbed very efficiently. Cadmium is likely to be in free ionic form in fresh water, whereas in foods, including foods of animal origin, it generally exists in a complex with a variety of ligands, including proteins such as metallothionein (NRC, 2005). The bioavailability of Cd in animal tissues is usually less than for cadmium chloride. However, the bioavailability of Cd in foods of plant origin may be somewhat higher than that of Cd salts (Prankel et al., 2004).

Animals with a high rate of feed intake appear to be affected at lower levels of dietary Cd than animals with lower feed intake. Thus, small animals with relatively high intake compared with their body size may be more sensitive to Cd effects than larger animals (NRC, 2005).

The toxicity of Cd is also affected by the nutritional and physiological state of an animal. Deficiencies in Zn, Fe, Cu, Ca, or protein increase the tissue accumulation and toxicity of Cd,

and vice versa (NRC, 2005). Cadmium has been shown to be absorbed at higher rates in immature animals than in adults (NRC, 2005).

Although Cd accumulation is greatest in the kidney, followed by liver, testes, pancreas and spleen, lower levels are also found in muscle and bone. The transfer of Cd to eggs and milk is very inefficient. Regardless of the level of Cd fed to animals, the concentration of Cd in meat, milk, and eggs is always lower, on a DW basis, than the level in the diet that animal consumed (NRC, 2005).

The maximum tolerable level of Cd is the dietary level that, when fed for a defined period of time, will not impair accepted indices of animal health or performance (NRC, 2005). Animals are able to tolerate acute exposure to 25 mg kg<sup>-1</sup> Cd in the diet for a few days (NRC, 2005). Functional indications of toxicosis in rodents, such as increased blood pressure, have been shown after a Cd intake of 1 mg kg<sup>-1</sup> diet for several months (NRC, 2005). A chronic dietary level of 10 mg kg<sup>-1</sup> is tolerated by poultry and livestock species. However, this level results in unacceptable levels in kidneys and liver, and, in some cases, also in muscles (NRC, 2005). Dogs can also tolerate 10 mg Cd kg<sup>-1</sup> over several years (NRC, 2005).

The toxicity of Cd in water appears to be very similar to that in feed, and the total intake from both sources should be considered (NRC, 2005).

The upper limit for Cd in complete feeds for animals at 1 mg kg<sup>-1</sup> diet has been set by WHO to protect consumers of animal products. In a review of potential contaminants in livestock feeds and proposed guideline levels for feed, MacLachlan et al. (2013) commented that advice on the maximum levels of exposure to Cd are challenging to develop. This is due to the accumulation of Cd with increasing duration of exposure, and slaughter of livestock usually occurs before steady-state concentrations are archived in tissues. Prankel et al. (2004) conducted a meta-analysis of feeding trials investigating Cd accumulation in livers and kidneys of sheep, and suggested that preventing livers and kidneys of older animals from entering the human food chain would be an appropriate measure to decrease the risk from Cd in foods of animal origin. Prankel et al. (2004) suggested that sheep fed a concentration of Cd from plant origin at 1 mg kg<sup>-1</sup> total feed DW (maximal limit in feed) would result in maximum residue levels in liver (0.5 mg kg<sup>-1</sup> wet weight) at approximately 200 days of exposure. The corresponding maximum residue level in kidney (1.0 mg kg<sup>-1</sup> wet weight) would be reached at approximately 150 days.

#### 7.3.3 Dietary human exposure

#### 7.3.3.1 Tolerable weekly intake

Diet is the main source of Cd exposure in the non-smoking general population (EFSA, 2009). The absorption of Cd is low (3 - 5%), but it accumulates efficiently in liver and kidney (EFSA, 2009; VKM, 2015). Cadmium is taken up in bone during growth and remodelling

(VKM, 2015). The biological half-life of Cd is long, 10 to 30 years in healthy humans (EFSA, 2009; VKM, 2012).

EFSA established a tolerable weekly intake (TWI) for Cd based on an increased risk of reduced kidney function in adults following long-term dietary exposure (EFSA, 2009). The TWI was set at  $2.5 \mu g$  Cd  $kg^{-1}$  BW per week (EFSA, 2009).

The TWI value was set on the basis of a meta-analysis of published data on the dose-response relationship between urinary Cd concentrations and the renal biomarker beta-2-microtubulin (B2M), an indicator of Cd-induced tubular damage (EFSA, 2009). A BMDL5 (benchmark dose lower confidence limit for a 5 % increase in the prevalence of elevated B2M) of 4  $\mu$ g Cd g<sup>-1</sup> creatinine was derived. To account for inter-individual variation of Cd concentrations in urine, an adjustment factor of 3.9 was applied, leading to a value of 1.0  $\mu$ g Cd g<sup>-1</sup> creatinine. One-compartment modelling was then used to estimate the relationship between urinary Cd concentrations and dietary Cd exposure. The model estimated a dietary Cd exposure leading to a critical urinary Cd concentration of 1  $\mu$ g Cd g<sup>-1</sup> creatinine after 50 years of exposure. For 95% of the population, this exposure was estimated as 0.36  $\mu$ g Cd kg<sup>-1</sup> BW per day, corresponding to 2.52  $\mu$ g Cd kg<sup>-1</sup> BW per week. Due to the long half-life of Cd in the human body, EFSA stated that the TWI should be set on a weekly basis, rather than a daily basis. As the data used in the modelling was related to an early indicator of renal changes, no further adjustment or uncertainty factors were applied, and the TWI was set at 2.5  $\mu$ g Cd kg<sup>-1</sup> BW per week.

In 2010, the Joint FAO/WHO Expert Committee on Food Additives (JECFA) established a PTMI (provisional tolerable monthly intake) of 25 µg kg<sup>-1</sup> BW per month (FAO/WHO, 2010), corresponding to a weekly intake of approximately 5.8 µg Cd kg<sup>-1</sup> BW. Following the JECFA evaluation, EFSA compared the two approaches (EFSA, 2011). The same dataset and two primary components (the use of a concentration-effect model and a toxicokinetic model) were used in the assessments. However, there were some methodological differences between the assessments; identification of reference point, statistical method used for evaluation of variability and uncertainty of biomarkers, and the method used for transformation of urinary Cd concentrations to dietary intake of Cd. EFSA found that their approach was appropriate and reaffirmed the TWI of 2.5 µg Cd kg<sup>-1</sup> BW per week (EFSA, 2011). VKM agrees with the EFSA approach and uses a TWI of 2.5 µg kg<sup>-1</sup> BW per week.

#### 7.3.3.2 Dietary exposure to cadmium

VKM concluded in their risk assessment of dietary Cd exposure in the Norwegian population, that exposure of the Norwegian adult population to Cd is comparable to the exposure of the adult European population (VKM, 2015). Long-term exposure above the TWI from a regular diet is unlikely in Norwegian adults, but additional consumption of foods with a high Cd content, e.g., crab and fish liver, may lead to exposure exceeding the TWI (VKM, 2015).

In 2012, EFSA estimated the dietary Cd exposure in the European population (EFSA, 2012). The estimated exposure is summarised in Table 7.3.3.2-1. For adolescents, adults, the

elderly and the very elderly, the mean middle bound (MB) exposures ranged from 1.63 to 2.20  $\mu$ g Cd kg<sup>-1</sup> BW per week; all exposures were below the TWI. Infants, toddlers, and other children had exposures (mean MB exposures of 2.74 to 3.96  $\mu$ g Cd kg<sup>-1</sup> BW per week) exceeding the TWI of 2.5  $\mu$ g Cd kg<sup>-1</sup> BW per week. Weighing the estimated exposures for each group by the percentage of the number of years they contribute to an average lifetime of 77 years, an average lifetime exposure was estimated. The average MB lifetime dietary exposure to Cd was calculated to be 2.04  $\mu$ g Cd kg<sup>-1</sup> BW per week.

**Table 7.3.3.2-1.** Middle bound (MB) and 95-percentile (P95) dietary Cd exposure in μg kg<sup>-1</sup> BW per week for each age group and as a mean and 95-percentile average lifetime exposure calculated by weighting the contribution of each age group according to the number of years covered (different range of countries covered in the respective age groups) (EFSA, 2012).

Age group	N	Mean MB	P95 MB
Infants	876	2.74	6.56
Toddlers	1597	4.85	8.19
Other children	8468	3.96	6.58
Adolescents	6329	2.2	4.17
Adults	30788	1.7	3.09
Elderly	4056	1.56	2.82
Very elderly	1614	1.63	2.87
Adjusted average <sup>1</sup>		2.04	3.66

<sup>&</sup>lt;sup>1</sup>The age groups represent 1 year for infants (1.3%), 2 years for toddlers (2.6%), 7 years for other children (9.1%), 8 years for adolescents (10.4%), 47 years for adults (61%), 10 years for the elderly (13.0%), and 2 years for the very elderly (2.6%) (Source: EFSA, 2012).

Dietary Cd exposure has been estimated in Denmark and Sweden. The mean daily exposure among Danish children (age 4 to 14 years) was 0.31  $\mu$ g Cd kg<sup>-1</sup> BW (DTU, 2013), corresponding to 2.17  $\mu$ g Cd kg<sup>-1</sup> BW per week. Among Danish adults (15 to 75 years of age), the estimated exposure was 0.15  $\mu$ g Cd kg<sup>-1</sup> BW per day (DTU, 2013), corresponding to 1.05  $\mu$ g Cd kg<sup>-1</sup> BW per week. In Sweden, the median exposures were 0.75 and 0.79  $\mu$ g Cd kg<sup>-1</sup> BW per week for women and men, respectively (SLV, 2017).

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#### 7.3.3.3 Food groups contributing to cadmium exposure

In Europe, grains and grain products, vegetables and vegetable products, and starchy root tubers are the food groups that contribute most to dietary Cd exposure (EFSA, 2012). At a more detailed level, the foods contributing the most to the dietary Cd exposure across all age groups are potatoes, bread and rolls, fine bakery wares, chocolate products, leafy vegetables, and water molluscs.

In Denmark and Sweden, cereals, potatoes and vegetables contribute the most to dietary Cd exposure.

# 8 Exposure assessment

# 8.1 Modelling environmental concentration soil (PEC<sub>soil</sub>)

Predicted soil concentrations, PEC<sub>soil</sub>, over time have been calculated using equation (8) (Amundsen et al., 2000).

$$Cd_{s}(t) = Cd_{s}(0) \cdot e^{-(k_{p}+k_{l})t} + (\frac{k_{l}}{k_{p}+k_{l}}) \cdot (1 - e^{-(k_{p}+k_{l})t})$$
 (Eq. 8)

Where

Cd<sub>s</sub> = cadmium concentration in soil at time t (mg kg<sup>-1</sup> DW)

 $Cd_s(0)$  = present concentration in soil (mg kg<sup>-1</sup> DW)

 $K_i = k_{input}$ , input rate of Cd (yr<sup>-1</sup>)

 $K_p = K_{plant-removal}$ , Cd removal by plants (yr<sup>-1</sup>) (Eq. 5)

 $K_l = K_{leaching}$ , Cd leaching from the plough layer (yr<sup>-1</sup>) (Eq. 2)

This equation is based on Cd input and Cd loss kinetic, removal rate of Cd via uptake in plants that are harvested and removed from the field, and leachates from the field per year.

The input parameters used for each region for predicting PEC<sub>soil</sub> are shown in Table 8.1-1. A soil depth of 0.2 m (Eq. 2) is used in ECHA (2016) and selected because this range usually has a high root density of crops and represents the ploughing depth.

For each of the four municipalities evaluated, different scenarios for crop rotations, reflecting the most common rotation for that municipality or district, were used for estimating the removal at each location (Table 5-1).

In calculating the Cd concentration in soil over a long timespan (100 years), the removal of Cd from the soil through plant uptake and harvesting, commonly termed output via crop offtake, has been taken into account (section 6.2.2 and equation 5).

Predicted soil concentrations (PEC<sub>soil</sub>) over time are shown in Table 8.1-2 and in Fig. 8.1-1. For all cases, except at Ås, the removal rate from soil, k ( $\sum k_{leaching}$ ,  $k_{plant-removal}$ ), was higher than Cd input rate ( $k_{input}$ ,  $\sum$ mineral P fertilisers, lime products, sewage sludge, and atmospheric deposition). At the locations Stange, Melhus and Time, it was predicted a decrease of Cd concentration in soil over time for all evaluated scenarios, including maximal allowed sewages sludge (only included in Stange) and use of mineral P fertiliser with ML 137.4 mg Cd  $kg^{-1}$  P (corresponding to 60 mg Cd  $kg^{-1}$  P<sub>2</sub>O<sub>5</sub>). The percent reduction over time varied at the different cases.

**Table 8.1-1.** Summary of soil quality, and precipitation and infiltration parameters used for estimating soil Cd concentration over time, leaching, runoff and transfer to crops and forage.

	County		South- eastern	Hedmark	South- western	Trøndelag
	Municipality		Ås	Stange	Time	Melhus
	Cd	mg kg	0.28	1.70	0.22	0.11
	background	DW <sup>-1</sup>				
	pH (CaCl <sub>2</sub> )		5.6	5.6	5.3	5.7
	SOM	%	5.7	6.7	4.1	6.3
Soil quality	OC	%	3.4	4.0	2.4	3.7
Son quanty	Clay content	%	22.6	15.5	8.2	17.0
	Soil dry	kg L <sup>-1</sup>	1.26	1.26	0.81	1.20
	density					
	K <sub>dsoil</sub>	L kg <sup>-1</sup>	217.6	247.4	115.9	257.8
	K <sub>soil-water</sub>	m <sup>3</sup> m <sup>-3</sup>	274	312	94	310
Precipitation	Precipitation <sup>1</sup>	mm yr <sup>-1</sup>	672	672	1464	1236
and	Rate of	Fraction	0.4	0.4	0.7	0.7
infiltration	infiltration <sup>1</sup>	(F)				

<sup>&</sup>lt;sup>1</sup> Precipitation (annual mean precipitation in the period 2003-2012) and infiltration parameters used in VKM (2014) were applied, and it was chosen the same parameter values for Ås as for Stange.

In a 10- year perspective, the reduction of Cd in soil was in the range of 3-10% for all ML of Cd in mineral fertiliser at Stange and Melhus (all crop rotations), while at Time it was around 40%. At Ås with application of maximal allowed sewages sludge and use of mineral P fertiliser with ML at 137.4 and 91.6 mg Cd kg $^{-1}$  P, the predicted soil concentration (PEC $_{soil}$ ) was slightly higher (0.283-0.286 mg Cd kg $^{-1}$  DW) than today practise and use of fertiliser with ML at 45.8 mg Cd kg $^{-1}$  P (0.280-0.281 mg Cd kg $^{-1}$  DW). This corresponded to a small increase, around 0.5-1.5%, of Cd in soil with use of ML at 137.4 and 91.6 mg Cd kg $^{-1}$  P.

Calculations based on of average application of sewage sludge as shown in data from SSB, rather than maximum allowed application, give no accumulation of Cd in soil over time, even with use of fertilisers with the highest ML at 137.4 mg Cd kg<sup>-1</sup> P at Ås (Table 8.1-3 and Fig. 8.1-2).

<u>In a 100-year perspective</u>, the estimated reduction increased to around 27-37% at Stange (both cereals and mixed crop production, however, mixed crop production has slightly higher reduction than cereals only), around 50-60% at Melhus with grass production, and around 90-95% at Time with grass production. The high removal rate of Cd at Time is also seen in Fig. 8.1-1. At Melhus, with cultivation of cereals (crop rotation barley and oat) the predicted Cd removal would decrease from 57% with today's practise and use of fertiliser to 28% with mineral P with ML 137.4 mg Cd kg<sup>-1</sup> P. A change in ML value of Cd had relative low influence on the removal of Cd over time at the other cases. At Stange and crop rotation with cereals, the predicted Cd concentration after 100 years and application of sewage sludge based on numbers from SSB, is slightly lower than after application of maximum allowed according to the regulation, 1.158 versus 1.233 mg Cd kg kg-1 DW (Table 8.1-2 and 8.1-3, Fig. 8.1-2),

resulting in a slightly higher reduction over time (e.g., for ML 137.4 mg Cd kg<sup>-1</sup> P 27.5% versus 31.9%).

In a 100-year perspective at Ås, the predicted Cd concentration increased to 0.294 mg Cd  $kg^{-1}$  DW and 0.315 mg Cd  $kg^{-1}$  DW, with ML at 91.6 and 137.4 mg Cd  $kg^{-1}$  P, corresponding to 4.2 and 11.7 % increase respectively.

Comparing PEC<sub>soil</sub> Cd concentration at 100 year after use of fertiliser with ML 137.4 mg Cd  $kg^{-1}$  P (0.315 mg Cd  $kg^{-1}$  DW) with soil concentration after use of today's fertiliser and based on the measured Cd concentration (NFSA, 2017) (0.263 mg Cd  $kg^{-1}$  DW), shows a 20% increase.

The contribution of Cd from fertilisers has a small effect on the PEC<sub>soil</sub> initially, but after 100 years, an increase in Cd levels in fertilisers (from the present level to 137.4 mg Cd kg<sup>-1</sup> P), may cause an increase in PEC<sub>soil</sub> of as much as 68% in Melhus, 50% in Time, 20% in Ås, and only 5-6% in Stange, where the background concentration in soil is high.

In previous Norwegian environmental and health risk assessments of Cd (and other as well), a default Kd value at 500 L kg<sup>-1</sup> (VKM, 2009) and an algorithm developed by McBride et al. (1977) (Amundsen et al. 2000) were used. The default value at 500 L kg<sup>-1</sup> is more than 4 times higher than the Kd used at Time and around 2 times higher than the Kd-values used at the other locations, and thus, predicted a higher sorption to soil. Use of the algorithm by McBride et al. (1977) also predicted a higher sorption of Cd to soil (see discussion Appendix III). These predictions are now assumed to be too high, and they might have caused in an overestimation of Cd concentrations in soil and underestimation of Cd concentrations in soil solutions and leachates from soil. An overestimates of soil concentrations will also result in overestimating the exposure of farm animals, and thereby humans, via their diet (e.g., grass, cereals, leafy and root vegetables). On the other hand, it will also underestimate the transfer from soil to surface water and groundwater.

The estimated higher leaching and reduction of Cd concentration in soil at several locations in the present risk assessment is the same trend as reported by Six and Smolders (2014) and others (e.g. SCAHT, 2015).

**Table 8.1-2.** Cd content in soil (PEC<sub>soil</sub>), presented as mg Cd kg<sup>-1</sup> DW, over time after application of mineral P fertiliser with ML Cd content of 45.8, 91.6, and 137.4 mg Cd kg<sup>-1</sup> P; corresponding to 20, 40, and 60 mg Cd kg<sup>-1</sup>  $P_2O_5$ , maximum allowed amount sewage sludge (40 tonnes ha<sup>-1</sup> yr<sup>-1</sup>) representing a realistic worst-case scenario, lime products and manure. For present practice, Cd concentration in the range of 25-50 mg Cd kg<sup>-1</sup> P is used. Current fertiliser practices are based on information from the local agricultural extension service (see Table 5-1), and Cd contribution is shown in Table 7.1.3-1. Numbers in bold are soil concentration with predicted accumulation of Cd in soil over time.

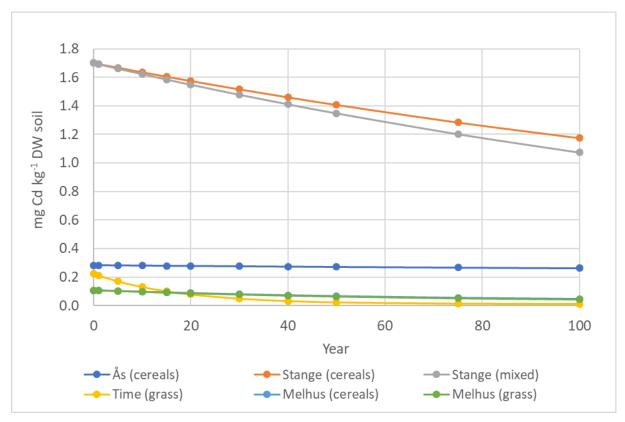
		Cd content of min	eral fertili	ser (mg C	d kg <sup>-1</sup> P)
		Current fertiliser	1		
		practices	45.8	91.6	137.4
Year					
Present - background	Ås (cereals)	0.282	0.282	0.282	0.282
1	As (Cereais)	0.282	0.282	0.282	0.282
10		0.280	0.281	0.283	0.286
100		0.263	0.273	0.294	0.315
Present - background	Character (companie)	1.700	1.700	1.700	1.700
1	Stange (cereals)	1.693	1.694	1.694	1.694
10		1.636	1.637	1.640	1.643
100		1.174	1.185	1.209	1.233
Present - background	Stange (potato,	1.700	1.700	1.700	1.700
1	carrot, cereals)	1.692	1.692	1.693	1.693
10		1.623	1.623	1.627	1.632
100		1.075	1.079	1.113	1.148
Present - background	Time (	0.224	0.224	0.224	0.224
1	Time (grass)	0.212	0.212	0.212	0.212
10		0.130	0.131	0.132	0.133
100		0.012	0.013	0.015	0.018
Present - background		0.108	0.108	0.108	0.108
1	Melhus (cereals)	0.107	0.107	0.107	0.107
10		0.097	0.098	0.100	0.103
100		0.046	0.052	0.065	0.078
Present - background	Mallana (	0.108	0.108	0.108	0.108
1	Melhus (grass)	0.107	0.107	0.107	0.107
10		0.097	0.097	0.098	0.099
100		0.044	0.045	0.051	0.056

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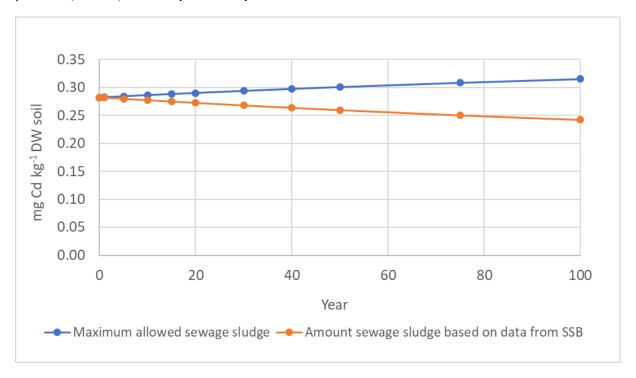
**Table 8.1-3.** Cd content in soil (PEC<sub>soil</sub>), presented as mg Cd kg $^{-1}$  DW, over time after application of mineral P fertiliser with ML Cd content of 45.8, 91.6, and 137.4 mg Cd kg $^{-1}$  P; corresponding to 20, 40, and 60 mg Cd kg $^{-1}$  P $_2$ O $_5$ , amount sewage sludge based on data from SSB, lime products and manure. For present practice, Cd concentration in the range of 25-50 mg Cd kg $^{-1}$  P is used. Current fertiliser practices are based on information from the local agricultural extension service (see Table 5-1), and Cd contribution is shown in Table 7.1.3-1.

		Cd content of min	eral fertili	ser (mg Co	d kg <sup>-1</sup> P)
		Current fertiliser	4		
		practices	45.8	91.6	137.4
Year					
Present - background	Ås (cereals)	0.282	0.282	0.282	0.282
1	AS (Cereais)	0.281	0.281	0.281	0.281
10		0.271	0.272	0.274	0.277
100		0.190	0.200	0.221	0.242
Present - background	Characa (22,000-12)	1.700	1.700	1.700	1.700
1	Stange (cereals)	1.693	1.693	1.693	1.693
10		1.627	1.628	1.631	1.634
100		1.098	1.109	1.133	1.158
Present - background	Stange (potato,	1.700	1.700	1.700	1.700
1	carrot, cereals)	1.692	1.692	1.693	1.693
10		1.623	1.623	1.627	1.632
100		1.075	1.079	1.113	1.148
Present - background	T: ( )	0.224	0.108	0.224	0.224
1	Time (grass)	0.212	0.212	0.212	0.212
10		0.130	0.131	0.132	0.133
100		0.012	0.013	0.015	0.018
Present - background		0.108	0.108	0.108	0.108
1	Melhus (cereals)	0.107	0.107	0.107	0.108
10		0.097	0.099	0.102	0.105
100		0.046	0.056	0.074	0.091
Present - background		0.108	0.108	0.108	0.108
1	Melhus (grass)	0.107	0.107	0.107	0.107
10		0.097	0.097	0.098	0.099
100		0.044	0.045	0.051	0.056

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**Fig. 8.1-1.** Predicted Cd concentration (mg Cd kg DW $^{-1}$ ) in soil over 100 years, using  $k_{leaching}$  equation 2, and with maximal allowed use of sewage sludge. Stange mixed had crop rotation potatoes, carrot, cereals (Table 5-1).



**Fig. 8.1-2.** Predicted Cd concentration (mg Cd  $kg^{-1}$  DW) in soil at Ås over 100 years, using  $k_{leaching}$  equation 2, and with maximum allowed used of sewage sludge and average amount based on data from SSB.

# 8.2 Modelling environmental concentrations in surface water (PEC<sub>surface water</sub>) and sediment (PEC<sub>sediment</sub>)

#### 8.2.1 Transport of Cd with drainage water

In the calculation of PEC<sub>soil</sub> described in section 7.4.1, it is assumed that precipitation excess infiltrates the soil and leaves the upper 20 cm of the soil profile containing Cd that has been leached from the soil. This is a simplified model of the very complex interaction between water and soil, where some of the water, especially during heavy rainfall, may be transported along the surface as runoff, and some of the water may bypass the upper soil layer as macropore flow through cracks or tunnels made by animals. These processes will tend to reduce the interaction of water with the soil and, thereby, affect the leaching of Cd. Here, the simplified assumption is that all the precipitation excess interacts fully with the soil resulting in a Cd concentration that is in equilibrium with the soil. This is a conservative approach regarding effect in the aquatic risk assessment.

After penetration of the upper soil profile, the excess precipitation may be transported via lateral flow or drainage pipes to surface water or may reach the groundwater. Here, the assumption is made that all the precipitation excess reaches a surface-water recipient. This is a worst-case scenario that is also conservative regarding the aquatic risk assessment.

#### 8.2.2 Predicted concentrations of Cd in surface water.

The predicted concentration of Cd in surface water has been calculated for a scenario in which the drainage water reaches a small stream with a constant dilution ratio, in accordance with the recommendation for local exposure estimation in TGD (ECB, 2003). The calculation involves removal from the aqueous medium by adsorption to suspended matter. The concentration in the local receiving water is calculated using the following equation:

$$PEC_{sw} = PEC_{regional} + \left(\frac{C_{effluent}}{(1 + Kp_{susp} \times SUSP_{water} \times 10^{-6}) \times DILUTION}\right)$$
 (Eq. 9)

Where:

PEC<sub>SW</sub> concentration of Cd in surface water (μg L<sup>-1</sup>)

PEC<sub>regional</sub> concentration of Cd in dilution water (μg L<sup>-1</sup>)

Ceffluent concentration of Cd in effluent water (drainage + runoff), (µg L<sup>-1</sup>)

Kp<sub>susp</sub> solids-water partitioning coefficient of suspended matter L kg<sup>-1</sup>

SUSP<sub>water</sub> concentration of suspended matter in the river (mg kg<sup>-1</sup>)

*DILUTION* dilution factor (default = 10)

A dilution factor of 10, which is the recommended default dilution for local PEC calculation in (ECB, 2003; ECHA, 2016), is used for calculation of  $PEC_{SW}$ .

For background concentration of Cd, the annual mean value obtained from major rivers in the regions where the case sites are located is used, as shown in Table 8.2.2-1 (NEA, 2018).

**Table 8.2.2-1.** Concentrations of Cd in regional rivers (5-year averages 2013-2017) used as  $C_{regional}$  for calculation of PEC<sub>SW</sub> (NEA, 2018).

Site	Regional river	PEC <sub>regional</sub> (μg Cd L <sup>-1</sup> )
Ås	Glomma	0.011
Stange	Glomma	0.011
Time	Orreelva	0.012
Melhus	Nidelva	0.003

A default value of 15 mg L<sup>-1</sup> is used as SUSP<sub>water</sub>, as recommended in the TGD (ECB, 2003; ECHA, 2016).

Kp<sub>susp</sub> for the suspended matter in the receiving water (130 000) is adopted from the European Cd risk assessment report (ECB, 2007).

The calculated PEC for surface-water recipients receiving effluents as drainage from soil are shown in Table 8.2.2-2.

**Table 8.2.2-2.** PEC<sub>SW</sub> for surface water recipients (µg Cd L<sup>-1</sup>) receiving effluents (eff.) from soil after application of mineral P fertilisers with the present, and alternative higher contents of Cd.

				mg C	Cd kg <sup>-1</sup> M	in.P ferti	iliser						
		Present		45.8		91.6		137.4					
Year		PECeff	PECsw	PECeff	PECsw	PECeff	PECsw	PECeff	PECsw				
			μg Cd L <sup>-1</sup>										
Background		1.296	0.055	1.296	0.055	1.296	0.055	1.296	0.055				
1	Ås	1.295	0.055	1.295	0.055	1.297	0.055	1.298	0.055				
10	AS	1.285	0.055	1.291	0.055	1.303	0.055	1.315	0.056				
100		1.211	0.052	1.254	0.054	1.351	0.057	1.447	0.060				
Background		6.871	0.244	6.871	0.244	6.871	0.244	6.871	0.244				
1	Stange	6.845	0.243	6.845	0.243	6.847	0.243	6.848	0.243				
10	(cereals)	6.612	0.235	6.617	0.235	6.629	0.236	6.641	0.236				
100		4.744	0.172	4.789	0.173	4.887	0.177	4.985	0.180				
Background	Stange	6.871	0.244	6.871	0.244	6.871	0.244	6.871	0.244				
1	(potato,	6.839	0.243	6.839	0.243	6.841	0.243	6.843	0.243				
10	carrot, cereals)	6.558	0.233	6.560	0.233	6.578	0.234	6.595	0.235				
100	cereais)	4.344	0.158	4.359	0.159	4.500	0.164	4.641	0.168				
Background		1.933	0.078	1.933	0.078	1.933	0.078	1.933	0.078				
1	Time	1.830	0.074	1.830	0.074	1.831	0.074	1.832	0.074				
10	Tille	1.126	0.050	1.129	0.050	1.139	0.051	1.148	0.051				
100		0.101	0.015	0.110	0.016	0.131	0.016	0.152	0.017				
Background		0.419	0.017	0.419	0.017	0.419	0.017	0.419	0.017				
1	Melhus	0.414	0.017	0.415	0.017	0.416	0.017	0.418	0.017				
10	(cereals)	0.377	0.016	0.384	0.016	0.396	0.016	0.407	0.017				
100		0.180	0.009	0.219	0.010	0.285	0.013	0.352	0.015				
Background		0.419	0.017	0.419	0.017	0.419	0.017	0.419	0.017				
1	Melhus	0.414	0.017	0.414	0.017	0.415	0.017	0.415	0.017				
10	(grass)	0.374	0.016	0.376	0.016	0.379	0.016	0.383	0.016				
100		0.170	0.009	0.176	0.009	0.197	0.010	0.217	0.010				

#### 8.2.3 Predicted concentrations of Cd in sediment

The concentration of Cd in the sediment of a recipient surface water can be calculated from the PEC<sub>SW</sub>, using the solid-water partition coefficient of sediments, Kp (EC, 2008) as:

$$PEC_{sediment} = Kp \times PEC_{sw}/1000$$
 (Eq. 10)

Where  $Kp = 9430 \text{ L kg}^{-1}$ , as calculated from measured average concentrations in European water and sediments in the European Cd risk assessment report (ECB, 2007).

The calculated PEC for surface water sediments receiving effluents from soil with the present background concentration of Cd are shown in Table 8.2.3-1.

**Table 8.2.3-1.** PEC for surface water sediments in recipients receiving effluents from soil after application of mineral P fertilisers with the present, and alternative higher, content of Cd.

		m	g Cd kg <sup>-1</sup> Mir	n.P fertiliser	
		Present	45.8	91.6	137.4
			mg Cd	kg <sup>-1</sup>	
Year					
Background		0.52	0.52	0.52	0.52
1	Ås	0.52	0.52	0.52	0.52
10	A5	0.51	0.52	0.52	0.52
100		0.49	0.50	0.54	0.57
Background		2.30	2.30	2.30	2.30
1	Stange	2.29	2.29	2.29	2.29
10	(cereals)	2.22	2.22	2.22	2.23
100		1.62	1.63	1.67	1.70
Background	Stange (potato,	2.30	2.30	2.30	2.30
1		2.29	2.29	2.29	2.29
10	carrot, cereals)	2.20	2.20	2.21	2.21
100		1.49	1.50	1.54	1.59
Background		0.73	0.73	0.73	0.73
1	Time	0.70	0.70	0.70	0.70
10		0.47	0.47	0.48	0.48
100		0.15	0.15	0.15	0.16
Background		0.16	0.16	0.16	0.16
1	Melhus	0.16	0.16	0.16	0.16
10	(cereals)	0.15	0.15	0.15	0.16
100	-	0.09	0.10	0.12	0.14
Background		0.16	0.16	0.16	0.16
1	Molhus (avass)	0.16	0.16	0.16	0.16
10	Melhus (grass)	0.15	0.15	0.15	0.15
100		0.08	0.08	0.09	0.10

## 8.2.4 Measured concentrations of Cd in surface waters

A survey of metal concentrations in Nordic lakes indicated a median concentration at 0.012  $\mu g \ L^{-1}$  and the 90-percentile of 0.055  $\mu g \ L^{-1}$  (Skjelkvåle et al., 1999). Yearly monitoring of several large rivers is performed by the Norwegian River Monitoring Programme. The last

report showed that annual mean values in four rivers in Southern Norway ranged from 0.006 to 0.011  $\mu$ g L<sup>-1</sup> (NEA, 2018).

Smaller rivers and streams in agricultural areas are expected to show higher concentrations of Cd, but data on measured concentrations are sparse. In the Ås area, a small stream draining cultivated land (Skuterudbekken) was analysed for Cd in 2017. The catchment area of the stream is 4.5 km², of which 61% is cultivated. The concentrations varied from 0.014-0.039  $\mu$ g Cd L⁻¹ during the year (one outlier of 200  $\mu$ g L⁻¹ was excluded) (NEA, 2019). Analysis of water from two streams draining agricultural areas near Stange in 2010, showed maximum concentrations of 0.21  $\mu$ g Cd L⁻¹ (Vingerjessa upstream Løten) and 0.22  $\mu$ g L⁻¹ (Fura at Haugset), (NEA, 2019). These data support the calculated PECsW values for Ås and Stange shown in Table 8.2.2-2.

From Rogaland County, where Time is located, field data of Cd are available from the outlet of Figgjoelva, a river with a 220 km<sup>2</sup> catchment of which a large portion of the lower area is agricultural land. The average concentration of Cd in the years 2012-2014 was 0.018  $\mu$ g L<sup>-1</sup> (n= 20). The highest concentration during this period was 0.038  $\mu$ g Cd L<sup>-1</sup>, which is 50% lower than the predicted PNEC (NEA, 2019).

In the Melhus municipality, Cd concentrations were measured in two small streams in 2015, and were 0.007  $\mu$ g L<sup>-1</sup> in Langbekken and 0.012  $\mu$ g L<sup>-1</sup> in Ratbekken (NEA, 2019). These concentrations support the calculated PEC for Melhus (0.017  $\mu$ g L<sup>-1</sup>)

Concentrations of Cd in 235 Norwegian lake sediments were measured by Rognerud et. al (1999). Samples were taken from the surface of the sediment (0-0.5 cm) and at 30-50 cm depth. The median value of Cd in the surface layer was 0.86 mg  $\rm kg^{-1}$  and the 90-percentile was 2.62 mg  $\rm kg^{-1}$ .

In the Ås area, the metal content of sediments in Lake Årungen has been measured by Zambon (2010). The catchment area of the lake is 52 km² and 53% of this area is cultivated. The average content of Cd in 192 samples of sediment at a depth of 2.5-5 cm was 0.56 mg Cd kg⁻¹, with a maximum of 1.5 mg Cd kg⁻¹. Riise et al. (2013) measured the vertical distribution of metals in sediment samples taken from the deepest part of Lake Årungen, and found that the concentration of Cd in the upper part of the profile was <0.4 mg Cd kg⁻¹. The concentration increased with depth, indicating that deposition of Cd has decreased over the last 40 years. The concentrations of Cd measured in Lake Årungen support the estimated PEC<sub>sediment</sub> for the Ås area.

From the shale area in Stange, sediment analysis has been reported from two streams; in Fura at Haugset, the concentration of Cd was 0.77 mg Cd kg<sup>-1</sup>, and in Vingerjessa upstream of Løten, sediment contained 0.36 mg kg<sup>-1</sup> (NEA, 2019). These concentrations are a factor of 3-6 times lower than the estimated PEC<sub>sediment</sub> for Stange.

# 8.3 Modelling concentrations in crops

Cereals, potatoes, carrots, and grass are the main food and forage crops. Uptake of Cd varies between plant species (discussed in section 6.2.2 and 7.2) and, based on the selected TFs, leafy vegetables and grass have the highest predicted Cd uptake (TF 0.36 g g<sup>-1</sup> DW based), followed by potato and carrot (TF 0.27 and 0.25 g g<sup>-1</sup> DW), with the lowest predicted uptake by cereals (TF 0.15 g g<sup>-1</sup> DW). Tables 8.3-1a and b show the predicted concentrations of Cd in feed and pasture plants, and plants for human consumption grown in the present agricultural soil. Predicted plant concentrations are related to TF and soil concentrations, and thus the highest predicted Cd concentration in dry weight is in potato and carrot grown on alum shale in Stange (Table 8.3-1a). The lowest Cd concentrations in DW is predicted for cereals grown in Melhus (Table 8.3-1b).

The predicted Cd concentration in plants corresponds the predicted trends in soil (Table 8.1-2 and 8.1-3): declining Cd concentrations in crops over time at Stange, Melhus and Time, and for crops produced in Ås, predicted reduction over time for current practice and ML 45.8 mg Cd kg<sup>-1</sup> P, even with use of maximum allowed sewage sludge application rate. However, with use of ML 91.6 and 137.4 mg Cd kg<sup>-1</sup> P, there is predicted an increase over time (Table 8.3-1b).

In evaluating the risk to humans, the exposure concentration is on a FW basis. The selected TF in FW are: cereals 0.14 g  $g^{-1}$  FW, leafy vegetables 0.108 g  $g^{-1}$  FW, potatoes 0.140 g  $g^{-1}$  FW, carrots 0.075 g  $g^{-1}$  FW, and grass 0.072 g  $g^{-1}$  FW.

On a FW basis, and taking into account water content, cereals are predicted to have the highest Cd content, with cereals grown in Stange (238  $\mu$ g kg<sup>-1</sup> FW) having around 6, 8, and 16 times higher concentrations of Cd than predicted for cereals grown in Ås, Time, and Melhus, respectively (Table 8.3-2). The predicted concentrations in potato and carrot grown in Stange were around 130  $\mu$ g kg<sup>-1</sup> FW. Based on the estimates and calculations performed in this risk assessment, the predicted Cd concentrations in FW in carrots, potatoes, and cereals grown in alum shale in Stange are above the Cd limit values (Table 3.3-1).

Compared with measured Cd values in crops grown on alum shale soil, the predicted concentrations in crops grown on alum shale soil at Stange were somewhat higher. Concentrations of Cd measured in cereals from Stange have been reported to be 84 and 187  $\mu$ g kg<sup>-1</sup> FW (n=2, converted from concentrations given in DW, Mellum et al., 1998; Esser, 1996), compared with the predicted concentrations of 283.4  $\mu$ g Cd kg<sup>-1</sup> FW. Similarly, measured concentrations in potatoes from Stange ranged from 19 to 65  $\mu$ g Cd kg<sup>-1</sup> FW (n=4) (Salbu et al., 2013) compared with the predicted value of 137  $\mu$ g Cd kg<sup>-1</sup> FW, and two analyses of carrot reported values of 90 and 95  $\mu$ g Cd kg<sup>-1</sup> FW (Salbu et al., 2013; Singh et al., 1995) compared with the predicted concentration 128  $\mu$ g Cd kg<sup>-1</sup> FW (Table 8.3-3). It has been reported that Cd concentration is generally higher in potato skin than its flesh. For example, Norton et al. (2015) found a significantly (P<0.001) more Cd in the skins of the baked potatoes compared to the flesh. On average the concentration of cadmium in the

potato skins was 23.7  $\mu$ g kg<sup>-1</sup> FW compared to 8.3  $\mu$ g kg<sup>-1</sup> FW in the flesh. Only one Cd analysis of salad grown on alum shale was found, and the concentration was reported to be 66  $\mu$ g Cd kg<sup>-1</sup> FW (Salbu et al., 2013).

Geogenic Cd in alum shale is known to have lower bioavailability than anthropogenic Cd; e.g., Kashem and Singh (2002) observed that TF decreased considerably from a naturally metal-rich alum shale soil compared with anthropogenically contaminated soils with tannery and city sewage input. The total Cd concentration was 2.5, 0.56, and 0.11 mg  $\rm kg^{-1}$  in alum shale, city sewage, and tannery soils, respectively, but the corresponding TF values for rice were 0.5, 1.1, and 1.8. In a sorption study of Cd with different soils, it was shown that adsorption of Cd in alum shale soil was higher than in sandy soil (Narwal and Singh, 1995). This implies a higher availability of Cd (and other metals) in anthropogenically contaminated soils.

Measured Cd values from crops grown at alum shale are scarce, and are difficult to interpret and evaluate against the predicted values. An ongoing 6-year Norwegian national project (2016-2021, funded by Norwegian Agriculture Agency), measures Cd (along with other trace elements and selected radionuclides) in crops grown on alum shale agricultural fields in Stange, and also investigates different measures to prevent Cd uptake in plants. The project will, among other, result in TFs for different crops, and also evaluate the effects of different measures at reducing plant uptake of Cd.

**Table 8.3-1a.** Estimated concentrations cadmium in mg kg<sup>-1</sup> DW in leafy vegetables, other vegetables (e.g., carrots), potatoes, cereals, and grass at Ås and Stange after application of mineral P fertiliser (today's use, and ML 45.8, 91.6 and 137.4 mg Cd kg<sup>-1</sup> P), manure, lime products and maximum allowed sewage sludge concentration.

Municipality (Crop Ro	otation)		Ås (cei	eal)			Stange (	(cereal)		Stange	(potato	-carrot-c	ereal)
Cd conc. in mineral P fertiliser (mg Cd kg <sup>-1</sup> P)		Present practice - Today's use <sup>1</sup>	45.8	91.6	137.4	Present practice - today's use <sup>1</sup>	45.8	91.6	137.4	Present practice - today's use <sup>1</sup>	45.8	91.6	137.4
Crop	Scenarios		1010	72.0	20711		1010	72.0	20211		1010	7210	
Leafy vegetables		0.102				0.612				0.612			
Other vegetables <sup>2</sup>		0.071				0.425				0.425			
Potatoes	Today	0.076				0.459				0.459			
Cereals		0.047				0.281				0.281			
Grass		0.102				0.612				0.612			
Leafy vegetables		0.101	0.101	0.102	0.102	0.610	0.610	0.610	0.610	0.609	0.609	0.609	0.609
Other vegetables <sup>2</sup>		0.070	0.070	0.071	0.071	0.423	0.423	0.423	0.424	0.423	0.423	0.423	0.423
Potatoes	1 yr	0.076	0.076	0.076	0.076	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457
Cereals		0.046	0.047	0.047	0.047	0.279	0.279	0.279	0.280	0.279	0.279	0.279	0.279
Grass		0.101	0.101	0.102	0.102	0.610	0.610	0.610	0.610	0.609	0.609	0.609	0.609
Leafy vegetables		0.101	0.101	0.102	0.103	0.589	0.589	0.590	0.592	0.584	0.584	0.586	0.587
Other vegetables <sup>2</sup>		0.070	0.070	0.071	0.072	0.409	0.409	0.410	0.411	0.406	0.406	0.407	0.408
Potatoes	<b>10</b> yr	0.076	0.076	0.077	0.077	0.442	0.442	0.443	0.444	0.438	0.438	0.439	0.441
Cereals		0.046	0.046	0.047	0.047	0.270	0.270	0.271	0.271	0.268	0.268	0.269	0.269
Grass		0.101	0.101	0.102	0.103	0.589	0.589	0.590	0.592	0.584	0.584	0.586	0.587
Leafy vegetables		0.095	0.098	0.106	0.113	0.423	0.427	0.435	0.444	0.387	0.388	0.401	0.413
Other vegetables <sup>2</sup>		0.066	0.068	0.073	0.079	0.293	0.296	0.302	0.308	0.269	0.270	0.278	0.287
Potatoes	100 yr	0.071	0.074	0.079	0.085	0.317	0.320	0.326	0.333	0.290	0.291	0.301	0.310
Cereals		0.043	0.045	0.048	0.052	0.194	0.195	0.199	0.203	0.177	0.178	0.184	0.189
Grass		0.095	0.098	0.106	0.113	0.423	0.427	0.435	0.444	0.387	0.388	0.401	0.413

<sup>&</sup>lt;sup>1</sup> Cd concentration in the range of 25 to 50 mg Cd kg<sup>-1</sup> P (Table 7.1.2.1.1-1), <sup>2</sup> e.g. carrots.

**Table 8.3-1b.** Estimated concentrations cadmium in mg kg<sup>-1</sup> DW in leafy vegetables, other vegetables (e.g. carrots), potatoes, cereals, and grass at Time and Melhus after application of mineral P fertiliser (today's use, and ML 45.8, 91.6 and 137.4 mg Cd kg<sup>-1</sup> P), manure, lime products and maximum allowed sewage sludge concentration.

Municipality			Time (g	ırass)			Melhus (	(cereal)			Melhus	(gras)	
Cd conc. in mineral P fertiliser (mg Cd kg <sup>-1</sup> DW)		Present practice - today's use <sup>1</sup>	45.8	91.6	137.4	Present practice - today's use <sup>1</sup>	45.8	91.6	137.4	Present practice - today's use <sup>1</sup>	45.8	91.6	137.4
Crop	Scenarios												
Leafy vegetables		0.081				0.039				0.039			
Other vegetables <sup>2</sup>		0.056				0.027				0.027			
Potatoes	Today	0.060				0.029				0.029			
Cereals		0.037				0.018				0.018			
Grass		0.081				0.039				0.039			
Leafy vegetables		0.076	0.076	0.076	0.076	0.038	0.039	0.039	0.039	0.038	0.038	0.038	0.039
Other vegetables <sup>2</sup>		0.053	0.053	0.053	0.053	0.027	0.027	0.027	0.027	0.027	0.027	0.027	0.027
Potatoes	1 yr	0.057	0.057	0.057	0.057	0.029	0.029	0.029	0.029	0.029	0.029	0.029	0.029
Cereals		0.035	0.035	0.035	0.035	0.018	0.018	0.018	0.018	0.018	0.018	0.018	0.018
Grass		0.076	0.076	0.076	0.076	0.038	0.039	0.039	0.039	0.038	0.038	0.038	0.039
Leafy vegetables		0.047	0.047	0.048	0.048	0.035	0.035	0.036	0.037	0.035	0.035	0.035	0.036
Other vegetables <sup>2</sup>		0.033	0.033	0.033	0.033	0.024	0.025	0.025	0.026	0.024	0.024	0.024	0.025
Potatoes	<b>10</b> yr	0.035	0.035	0.036	0.036	0.026	0.027	0.027	0.028	0.026	0.026	0.026	0.027
Cereals		0.022	0.022	0.022	0.022	0.016	0.016	0.017	0.017	0.016	0.016	0.016	0.016
Grass		0.047	0.047	0.048	0.048	0.035	0.035	0.036	0.037	0.035	0.035	0.035	0.036
Leafy vegetables		0.004	0.005	0.005	0.006	0.017	0.019	0.023	0.028	0.016	0.016	0.018	0.020
Other vegetables <sup>2</sup>		0.003	0.003	0.004	0.004	0.012	0.013	0.016	0.019	0.011	0.011	0.013	0.014
Potatoes	100 yr	0.003	0.003	0.004	0.005	0.013	0.014	0.018	0.021	0.012	0.012	0.014	0.015
Cereals		0.002	0.002	0.002	0.003	0.008	0.009	0.011	0.013	0.007	0.007	0.008	0.009
Grass		0.004	0.005	0.005	0.006	0.017	0.019	0.023	0.028	0.016	0.016	0.018	0.020

<sup>&</sup>lt;sup>1</sup> Cd concentration in the range of 25 to 50 mg Cd kg<sup>-1</sup> P (Table 7.1.2.1.1-1), <sup>2</sup> e.g. carrots.

**Table 8.3-2.** Predicted Cd concentrations in crops in this risk assessment with today's fertiliser practices, and ML values for Cd in selected crops (Table 3.3-1), all given as  $\mu g \ kg^{-1}$  FW.

	ML Cd in crops	Ås (cereals)	Stange (cereals)	Stange (potato-carrot- cereal)	Time (grass)	Melhus (cereal)	Melhus (grass)
Leafy vegetables		31	184	184	24	12	12
Other vegetables <sup>4</sup>	100	21	128	128	17	8	8
Potatoes	100 <sup>1</sup>	23	138	138	18	9	9
Cereals	100 <sup>2</sup> /200 <sup>3</sup>	40	238	238	31	15	15
Grass	1000	20	122	122	16	8	8

<sup>&</sup>lt;sup>1</sup> peeled potato, <sup>2</sup> barley and oat, <sup>3</sup> wheat, <sup>4</sup> e.g. carrots

**Table 8.3-3.** Measured Cd concentrations in crops, given as μg kg<sup>-1</sup> DW or μg kg<sup>-1</sup> FW, and related soils from field investigations, given as mg kg<sup>-1</sup> DW. Values in bold are data grown on alum shale in Hedmark region.

Crops	Cd in crops on alum shale	Cd in crops on alum shale	Cd in alum shale soil	Cd in crops in Swedish Soils
	(µg kg <sup>-1</sup> DW)	(μg kg <sup>-1</sup> FW)	(µg kg <sup>-1</sup> )	(µg kg <sup>-1</sup> DW)
Wheat	99 <sup>1</sup>	84 <sup>9</sup>	1000¹	69 <sup>5</sup>
Barley			10003¹	
Oats	220 <sup>3</sup>	1878	10003 <sup>1</sup>	315
Potato	1031	<b>19</b> <sup>6</sup> ; <b>32</b> <sup>6</sup> ; <b>65</b> <sup>6</sup> ; 30.9 <sup>9</sup>	9301	515
Carrot	3004	<b>95</b> <sup>6</sup> ; 90 <sup>7</sup>	930¹	2765
Grass	68 <sup>3</sup>	13.68	580 <sup>3</sup>	
Salad		66 <sup>6</sup>		
Cauliflower		<b>35</b> <sup>6</sup>		

Extracted from: ¹Mellum et al. (1998), ²Bærug and Singh (1990), ³Esser (1996), ⁴Singh et al. (1995), ⁵Ericsson et al. (1996), ⁶Salbu et al. (2013), ⁵Estimated values from ref⁴ on alum shale given as dry weight to fresh weight, <sup>8</sup>Estimated values from ref³ on alum shale.

# 8.4 Exposure of farm animals

For herbivorous livestock on pasture, such as cattle, sheep, goats, and horses, their whole ration may be comprised of pasture plants. However, dairy cows and goats usually receive grain-based feed (compound feed) also when at pasture, ranging from 0 to about 1/3 of total DW intake. According to Norwegian legislation, cows have to be kept outside for at least 8 weeks per year, and sheep and goats at least 16 weeks per year. Sheep, goats, and horses usually graze rough grass, but cattle may often graze on fertilised areas.

Animals at pasture usually also ingest some soil. Soil intake may depend on the pasture quality and the mineral needs of the animals. The intake of soil is supposed to constitute up to some percentage (5 %) of the DW ration.

Omnivorous animals, such as poultry and pigs, may ingest grass and other vegetables when outside. They may also ingest considerable amounts of soil and soil organisms, such as earthworms.

For herbivorous livestock, like cattle, sheep, goats, and horses, receiving feedstuff, the roughage constitutes a main part of the ration. In addition, compound feed (grain-based feed) or in some cases, potatoes etc. are given at a certain ratio (up to about 50 % of the DW ration to these species).

Small grains, oil seeds, an array of seed legumes, and some maize are common ingredients in pig feed and may be grown on fertilised soil. The main ingredients in poultry feeds are maize and small grains.

The normal amounts of dietary intake relative to the body weight of different livestock species at different physiological stages are shown in Table 8.4.-1. The table also shows the common composition of diets for grazing animals on pasture and fed at home.

**Table 8.4-1.** Dry weight (DW) intake of farm animals related to body weight and their relatively intake of compound feed, roughage and soil.

	Percent	Graz	ing animals		Fe	d animals	
	DW	Ratio of t	otal intake (	(DW)	Ratio of t	otal intake (	(DW)
Animal species and age group	intake related to body weight	Compound feed	Roughage	Soil	Compound feed	Roughage	Soil
Cattle							
Calves	3.0	0.10	0.85	0.05	0.25	0.75	0
Young heifers	2.7	0.10	0.85	0.05	0.25	0.75	0
Dry cows	1.7	0.10	0.85	0.05	0.15	0.85	0
High-lactation cows	4.0	0.50	0.45	0.05	0.5	0.50	0
Sheep							
Early weaned lambs	5.0	0	0.95	0.05	0.25	0.75	0
Finishing lambs	4.0	0	0.95	0.05	0.25	0.75	0
Adult sheep, maintenance	2.0	0	0.95	0.05	0.25	0.75	0
Adult sheep with twins	4.0	0	0.95	0.05	0.25	0.75	0
Goats							
Kids	3.5	0.10	0.85	0.05	0.25	0.75	0
Adult goats, maintenance	2.0	0.10	0.85	0.05	0.25	0.75	0
Adult lactating goats	6.0	0.50	0.45	0.05	0.50	0.50	0
Horses							
Adult maintenance	1.5	0.25	0.70	0.05	0.30	0.70	0
Mares in lactation	3.0	0.25	0.70	0.05	0.30	0.70	0
Pigs							
Piglets	10.0				1.00	0	0
Growing pigs	4.0				1.00	0	0
Adult pigs, maintenance	1.2				1.00	0	0
Lactating sow	3.2				1.00	0	0
Poultry					1.00		
Growing chickens	10.0				1.00	0	0
Laying hens	6.0				1.00	0	0
Broiler parents	6.0				1.00	0	0
Turkey	6.0				1.00	0	0

**Table 8.4.-2.** Predicted intake of Cd, given as μg Cd kg<sup>-1</sup> body weight (BW) per day (μg kg<sup>-1</sup> bw d<sup>-1</sup>) and mg Cd kg<sup>-1</sup> DW diet (mg kg<sup>-1</sup> dw diet), in ruminants and horses grazing and fed when housed before and after 100 years with use of mineral fertiliser (Min.P) containing maximum concentration of phosphorous (P).

		Time							Stange (cereals)							
		Gra	zing			Hou	sed			Gra	zing			Ηοι	ised	
	Cd 100 yr 134.7		yr 134.7	Cd 100 yr 134.7					Cd 100	yr 134.7			Cd 100	yr 134.7		
	Backgr		mg kg-1 Min.P		Background		mg kg <sup>-1</sup> Min.P		Background		mg kg <sup>-1</sup> Min.P		Background		mg kg <sup>-1</sup> Min.P	
	μg kg <sup>-1</sup>	mg kg <sup>-1</sup>	μg kg <sup>-1</sup>	mg kg <sup>-1</sup>	μg kg <sup>-1</sup>	mg kg <sup>-1</sup>	μg kg <sup>-1</sup>	mg kg <sup>-1</sup>	μg kg <sup>-1</sup>	mg kg <sup>-1</sup>	μg kg <sup>-1</sup>	mg kg <sup>-1</sup>	μg kg <sup>-1</sup>	mg kg <sup>-1</sup>	μg kg <sup>-1</sup>	mg kg <sup>-1</sup>
	bw d <sup>-1</sup>	dw diet	bw d⁻¹	dw diet	bw d <sup>-1</sup>	dw diet	bw d <sup>-1</sup>	dw diet	bw d⁻¹	dw diet	bw d⁻¹	dw diet	bw d <sup>-1</sup>	dw diet	bw d <sup>-1</sup>	dw diet
Cattle																
Calves	2.503	0.083	0.196	0.007	2.092	0.070	0.164	0.005	18.998	0.633	13.782	0.459	15.874	0.529	11.516	0.384
Young heifers	2.253	0.083	0.177	0.007	1.882	0.070	0.148	0.005	17.098	0.633	12.404	0.459	14.286	0.529	10.364	0.384
Dry cows	1.418	0.083	0.111	0.007	1.259	0.074	0.099	0.006	10.765	0.633	7.810	0.459	9.559	0.562	6.935	0.408
High-lactation																
cows	2.639	0.066	0.207	0.005	2.352	0.059	0.185	0.005	20.026	0.501	14.528	0.363	17.850	0.446	12.950	0.324
Sheep																
Early weaned																
lambs	4.390	0.088	0.345	0.007	3.486	0.070	0.274	0.005	33.320	0.666	24.173	0.483	26.456	0.529	19.193	0.384
Finishing lambs	3.512	0.088	0.276	0.007	2.789	0.070	0.219	0.005	26.656	0.666	19.338	0.483	21.165	0.529	15.355	0.384
Adult sheep,																
maintenance	1.756	0.088	0.138	0.007	1.394	0.070	0.109	0.005	13.328	0.666	9.669	0.483	10.583	0.529	7.677	0.384
Adult sheep																
with twins	3.512	0.088	0.276	0.007	2.789	0.070	0.219	0.005	26.656	0.666	19.338	0.483	21.165	0.529	15.355	0.384
Goats																
Kids	2.920	0.083	0.229	0.007	2.440	0.070	0.192	0.005	22.164	0.633	16.079	0.459	18.519	0.529	13.435	0.384
Adult goats,																
maintenance	1.669	0.083	0.131	0.007	1.394	0.070	0.109	0.005	12.952	0.648	9.188	0.459	10.583	0.529	7.677	0.384
Adult lactating																
goats	3.958	0.066	0.311	0.005	3.528	0.059	0.277	0.005	35.364	0.589	21.792	0.363	26.775	0.446	19.424	0.324
Horses																
Adult																
maintenance	1.153	0.077	0.091	0.006	1.013	0.068	0.080	0.005	8.753	0.584	6.350	0.423	7.688	0.513	5.578	0.372
Lactating mares	2.307	0.077	0.181	0.006	2.026	0.068	0.159	0.005	17.506	0.584	12.700	0.423	15.377	0.513	11.155	0.372

	Ås									Melhus (grass)							
		Gra	zing			Нои	ised			Gra	zing			Hou	ısed		
			Cd 100 y	yr 134,7	Cd 100 yr 134.7					Cd 100	yr 134.7			Cd 100 y	yr 134.7		
	Backg	round	mg kg-1 Min.P		Background		mg kg	mg kg <sup>-1</sup> Min.P		Background		mg kg <sup>-1</sup> Min.P		Background		mg kg <sup>-1</sup> Min.P	
	μg kg <sup>-1</sup>	mg kg <sup>-1</sup>	μg kg <sup>-1</sup>	mg kg <sup>-1</sup>	μg kg <sup>-1</sup>	mg kg <sup>-1</sup>	μg kg <sup>-1</sup>	mg kg <sup>-1</sup>	μg kg <sup>-1</sup>	mg kg <sup>-1</sup>							
	bw d <sup>-1</sup>	dw diet	bw d <sup>-1</sup>	dw diet	bw d <sup>-1</sup>	dw diet	bw d <sup>-1</sup>	dw diet	bw d⁻¹	dw diet							
Cattle																	
Calves	3.06	0.10	3.07	0.10	2.63	0.09	2.94	0.10	3.59	0.12	2.39	0.08	1.01	0.03	0.52	0.02	
Young heifers	2.76	0.10	2.77	0.10	2.37	0.09	2.65	0.10	3.94	0.15	2.15	0.08	0.91	0.03	0.47	0.02	
Dry cows	1.74	0.10	1.74	0.10	1.59	0.09	1.77	0.10	2.04	0.12	1.36	0.08	0.61	0.04	0.31	0.02	
High-lactation																	
cows	3.21	0.08	3.11	0.08	2.96	0.07	3.31	0.08	4.46	0.11	3.01	0.08	1.13	0.03	0.59	0.01	
Sheep																	
Early weaned																	
lambs	5.38	0.11	5.43	0.11	4.39	0.09	4.90	0.10	6.10	0.12	4.04	0.08	1.68	0.03	0.87	0.02	
Finishing																	
lambs	4.31	0.11	4.34	0.11	3.51	0.09	3.92	0.10	4.88	0.12	3.23	0.08	1.34	0.03	0.70	0.02	
Adult sheep,																	
maintenance	2.15	0.11	2.17	0.11	1.76	0.09	1.96	0.10	2.44	0.12	1.62	0.08	0.67	0.03	0.35	0.02	
Adult sheep																	
with twins	4.31	0.11	4.34	0.11	3.51	0.09	3.92	0.10	4.88	0.12	3.23	0.08	1.34	0.03	0.70	0.02	
Goats																	
Kids	3.58	0.10	3.59	0.10	3.07	0.09	3.43	0.10	4.19	0.12	2.79	0.08	1.18	0.03	0.61	0.02	
Adult goats,																	
maintenance	2.04	0.10	2.05	0.10	1.76	0.09	1.96	0.10	2.40	0.12	1.59	0.08	0.67	0.03	0.35	0.02	
Adult lactating																	
goats	4.81	0.08	4.67	0.08	4.44	0.07	4.96	0.08	6.68	0.11	4.52	0.08	1.70	0.03	0.88	0.01	
Horses																	
Adult																	
maintenance	1.41	0.09	1.40	0.09	1.28	0.09	1.42	0.09	1.75	0.12	1.17	0.08	0.49	0.03	0.25	0.02	
Lactating																	
mares	2.82	0.09	2.80	0.09	2.55	0.09	2.85	0.09	3.50	0.12	2.34	0.08	0.98	0.03	0.51	0.02	

In Table 8.4.-2 is shown predicted intake of Cd in ruminants and horses grazing and fed when housed before and after 100 years with use of fertiliser containing maximum concentration of P. If pigs and poultry are given a diet correspondingly grown at fertilized soil they will be similarly exposed to Cd. However, most pigs and poultry are fed commercial compound feed. Therefore, these species are not shown in this table. The most likely source of elevated Cd exposure for most animals kept in captivity is mineral supplements used in the feed. Phosphate and zinc sources, in particular, can be important contributors (NRC, 2005). Cadmium accumulates in plants grown in cadmium-rich soil – more so in plants grown in acid soils. In plants, Cd is concentrated in the leaves with lower levels in seeds and roots (He and Singh, 1994), so grazing animals are more exposed to the parts with higher levels.

In aquatic and benthic environments, Cd levels are relatively uniform throughout the food chain, usually without biomagnification (IPCS, 1992b).

The results of estimated concentrations in plants, feed and forage are expressed by DW.

The model for Cd concentrations in feed and pasture plants, even at the highest level of mineral fertiliser tested for 100 years (worst case), found that the Cd level would decline.

#### 8.5 Humans

The use of cadmium-containing fertilisers may affect human dietary exposure in several ways. Human exposure to Cd may increase due to higher concentrations in drinking water through runoff to drinking water sources, due to higher Cd concentrations in crop plants cultivated in fertiliser-treated soil, and through carry-over to animal-derived food products. Runoff from agricultural water is heavily diluted before reaching potential drinking water sources, and any change in human exposure to Cd via drinking water has not been included in the modelled scenarios.

Cd concentration in groundwater and in drinking water from private wells in alum shale areas might be higher than in other areas. The limit value for Cd in Norwegian drinking water regulation is 5  $\mu$ g L<sup>-1</sup>, however, the NFSA recommend drinking water not to contain > 3  $\mu$ g L<sup>-1</sup>, and WHO has a recommended health limit at 3  $\mu$ g L<sup>-1</sup>. As far as we know, there is no available data on Cd concentrations in drinking water from private drinking wells. Predicted pore water concentration based on Kd value for Cd in Stange (Kd 247.4 L kg<sup>-1</sup>) is 6.78  $\mu$ g L<sup>-1</sup>. However, it is reasons to believe the Kd for geogenic Cd in alum shale is higher, and, thus, the pore water might be lower. This is also of relevance for exposure of farm animals which gain drinking water from groundwater sources.

In animals, Cd is mainly deposited in the internal organs, and normally only very low levels are found in muscle. Offal consumption in Norway is low and the potential increase in liver concentrations of Cd would contribute little to total dietary exposure. This risk assessment therefore focuses on altered Cd exposure resulting from changes in Cd content of food-crop plants.

In 2015, VKM considered the possibility of estimating the dietary exposure to Cd in the Norwegian population, based on Norwegian Food Consumption data combined with Norwegian occurrence data, completed with occurrence data from EFSA for food items for which Norwegian data were lacking. However, VKM concluded that the uncertainties would be too high for the estimates to be helpful (VKM, 2015). This situation has not changed in recent years, as there are not many more available data from different food groups. VKM also reviewed available occurrence data in Norway and compared these with the EFSA data, and also compared available biomonitoring data from Norway, Sweden, and other countries; it was concluded that the median dietary exposures in Norway were similar to those of other countries (VKM, 2015). Based on these comparisons, VKM assessed the risk from dietary Cd, based on the average exposure estimated by EFSA, coupled with relevant Norwegian scenarios for intake of particular food items with high Cd concentrations that were not included in mean dietary estimates, such as brown crab meat, mussels, and fish liver (VKM, 2015). The mean dietary Cd exposures for children and adolescents, as estimated by EFSA and VKM, were in the range of the TDI or above, with the highest exposures estimated for children (EFSA, 2012; VKM, 2015).

The Cd concentrations in cereal products will not change significantly after 1 year of application of fertiliser in any scenario. After 10 years of application, the estimated Cd concentrations in cereals are estimated to decrease by approximately 40% in Time, 10-20% in Melhus, by 0-10% in Stange while no changes is expected at As. After 100 years, the concentrations are estimated to decrease by 30-40 % in Stange, about 30% in Melhus and by more than 90% in Time. In Ås, the scenarios vary from a decrease of 4 % to an increase of 11%, depending on Cd concentration in the fertiliser. Overall, the estimates show small differences between the selected Cd concentrations in the fertiliser (see Tables 8.3-1a and b). The previous EFSA and VKM estimates showed that Cd exposure from cereal and cereal products would contribute to a total of 25-36% of the estimated current mean exposures (EFSA 2012, VKM 2015). In a Norwegian study, cereal and cereal products contributed 31%, 44%, and 54% of the dietary Cd intake for the low, median, and high daily intake, respectively (Fange, 2005). The predicted reductions in cereal Cd concentrations could reduce the total dietary exposure. However, cereals from different regions and imported cereals are mixed in the mills during the production process, and therefore the final outcome would depend on the production of cereals in the different regions and the percentage of imported cereals. It is unlikely that persons are exposed to cereals grown only in the Stange area.

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After 1 year of application of fertiliser, the estimates indicate that there may be a 5% decrease in Cd concentrations in potatoes from the Time area, but no change in Cd concentrations in potatoes are expected in the other areas (Tables 8.3-1a and b). There are no differences between the different Cd concentrations in the fertiliser. After 10 years, the estimates indicate a 50% reduction in Cd concentrations in potatoes from Time, but the decrease is only 0-10% in the other regions, with only minor differences between the four different Cd concentrations in the fertiliser (Tables 8.3-1a and b). After 100 years, the concentrations of Cd in potatoes is estimated to decrease by 30-50% at Melhus and Stange and up to > 90% in Time. At As a fertiliser concentration of 137.4 Cd kg<sup>-1</sup> P is expected to increase the Cd concentrations in potatoes with 11%, while a decrease of 3% is expected using fertiliser containing 45.8 mg Cd kg<sup>-1</sup> P. The difference between continuing with the current practice and the three different scenarios for Cd concentrations are small (Tables 8.3-1a and b). Potatoes have been estimated to contribute to approximately 25% of the dietary Cd exposure in EU, Sweden, and Norway and a lower Cd concentration in potatoes would have a significant effect on the dietary Cd intake. An estimation of the reduction would require detailed information on the proportion of potatoes cultivated in the different soil types and Cd concentrations in imported potatoes. Potatoes and vegetables are generally distributed through commercial partners, where products from different producers are mixed and also imported potatoes are consumed. It is therefore unlikely that consumers will only purchase products grown on soil with high Cd concentrations. The exceptions may be for persons cultivating their own vegetables or frequently buying products directly from a farm in this area.

The models predict a slight increase in Cd concentrations in leafy vegetables (see Tables 8.3-1a and b). According to EFSA exposure estimates, leafy vegetables only contribute to 3.9% of the total dietary exposure (EFSA, 2012). A minor increase in Cd concentrations in leafy vegetables is therefore unlikely to have a large influence on the total dietary exposure to Cd.

According to Calabrese et al. (1989), 90% of the children ingested less than 0.2 g soil per day (Calabrese et al., 1989). This amount of soil was later used by the Norwegian Environment Agency to establish quality classes for soil in kindergardens and playing grounds in Norway (Alexander, 2006). VKM has therefore chosen to use 0.2 g soil/day as an amount a child could ingest of soil per day in this risk assessment. Consumption of 0.2 g soil with the present median soil concentration at Stange (1.7 mg Cd kg<sup>-1</sup>) would result in ingestion of 0.34  $\mu$ g day<sup>-1</sup> or 0.034  $\mu$ g kg<sup>-1</sup> BW for a child of 10 kg which is about 1 % of the dietary intake in infants estimated by EFSA (Table 7.3.3.2-1).

# 9 Risk characterisation

# 9.1 Risk characterisation in terrestrial organisms

The risk characterisation is based on the PEC<sub>soil</sub> as shown in Table 8.1-2, and the PNECs described in section 7.3.1.

#### 9.1.1 Direct exposure of terrestrial organisms

The RCRs shown in Table 9.1.1-1 are below 1 for all scenarios, indicating no risk from Cd for terrestrial organisms due to direct exposure to soil. The highest RCR (0.74) is found in Stange, where the present background Cd concentration is 26% below the PNEC $_{soil}$ . The PEC and RCR increase with the level of Cd in fertilisers at all locations/crops (horizontally in table). This is most obvious at sites with low background concentrations and high application rate of fertilisers, such as Melhus with cereal cultivation. For this scenario, the PEC and RCR after 100 years with the highest content of Cd in mineral P are 68% higher than with the present content of Cd in mineral P. With the exception of Ås, where the two highest levels of P in fertilisers cause an accumulation of Cd in soil, the PEC and RCR decline with time for all sites/crop cultivation scenarios.

**Table 9.1.1-1**. Predicted environmental concentrations (PEC, mg Cd  $kg^{-1}$  DW) and RCR for direct effects on terrestrial organisms (PNEC = 2.3 mg Cd  $kg^{-1}$ ).

		mg Cd kg <sup>-1</sup> Min.P fertiliser										
Year		Pres	ent	45	<b>5.8</b>	91.0	5	13	7.4			
		PEC	RCR	PEC	RCR	PEC	RCR	PEC	RCR			
Background		0.282	0.12	0.282	0.12	0.282	0.12	0.282	0.12			
1	Ås	0.282	0.12	0.282	0.12	0.282	0.12	0.282	0.12			
10	7.5	0.280	0.12	0.281	0.12	0.283	0.12	0.286	0.12			
100		0.263	0.11	0.273	0.12	0.294	0.13	0.315	0.14			
Background		1.700	0.74	1.700	0.74	1.700	0.74	1.700	0.74			
1	Stange	1.693	0.74	1.694	0.74	1.694	0.74	1.694	0.74			
10	(cereals)	1.636	0.71	1.637	0.71	1.640	0.71	1.643	0.71			
100		1.174	0.51	1.185	0.52	1.209	0.53	1.233	0.54			
Background	Stange (potato, carrot,	1.700	0.74	1.700	0.74	1.700	0.74	1.700	0.74			
1		1.692	0.74	1.692	0.74	1.693	0.74	1.693	0.74			
10		1.623	0.71	1.623	0.71	1.627	0.71	1.632	0.71			
100	cereals)	1.075	0.47	1.079	0.47	1.113	0.48	1.148	0.50			
Background		0.224	0.10	0.224	0.10	0.224	0.10	0.224	0.10			
1	Time	0.212	0.09	0.212	0.09	0.212	0.09	0.212	0.09			
10	Time	0.130	0.06	0.131	0.06	0.132	0.06	0.133	0.06			
100		0.012	0.01	0.013	0.01	0.015	0.01	0.018	0.01			
Background		0.108	0.05	0.108	0.05	0.108	0.05	0.108	0.05			
1	Melhus	0.107	0.05	0.107	0.05	0.107	0.05	0.108	0.05			
10	(cereals)	0.097	0.04	0.099	0.04	0.102	0.04	0.105	0.05			
100		0.046	0.02	0.056	0.02	0.074	0.03	0.091	0.04			
Background		0.108	0.05	0.108	0.05	0.108	0.05	0.108	0.05			
1	Melhus	0.107	0.05	0.107	0.05	0.107	0.05	0.107	0.05			
10	(grass)	0.097	0.04	0.097	0.04	0.098	0.04	0.099	0.04			
100		0.044	0.02	0.045	0.02	0.051	0.02	0.056	0.02			

# 9.1.2 Direct exposure of agricultural plants

The RCRs are below 1, indicating no risk from direct exposure to soil for terrestrial plants (Table 9.1.2-1). With the exception of Ås, where Cd accumulates in soil at the two highest levels of Cd in fertilisers, the PEC and RCR decrease with time.

**Table 9.1.2-1.** Predicted environmental concentrations (PEC, mg Cd kg<sup>-1</sup> DW) and RCRs for direct effects on higher plants (PNEC =  $2.8 \text{ mg Cd kg}^{-1}$ ), with application of mineral P fertilisers with the present, and alternative higher, content of Cd for 0-100 years.

				mg Co	d kg <sup>-1</sup> M	in.P fer	tiliser		
Year		Pres	ent	45	.8	91	6	137	<b>'.4</b>
		PEC	RCR	PEC	RCR	PEC	RCR	PEC	RCR
Background		0.282	0.10	0.282	0.10	0.282	0.10	0.282	0.10
1	Ås	0.282	0.10	0.282	0.10	0.282	0.10	0.282	0.10
10	, <b></b>	0.280	0.10	0.281	0.10	0.283	0.10	0.286	0.10
100		0.263	0.09	0.273	0.10	0.294	0.10	0.315	0.11
Background		1.700	0.61	1.700	0.61	1.700	0.61	1.700	0.61
1	Stange (cereals)	1.693	0.60	1.694	0.60	1.694	0.60	1.694	0.61
10		1.636	0.58	1.637	0.58	1.640	0.59	1.643	0.59
100		1.174	0.42	1.185	0.42	1.209	0.43	1.233	0.44
Background	Stange (potato, carrot,	1.700	0.61	1.700	0.61	1.700	0.61	1.700	0.61
1		1.692	0.60	1.692	0.60	1.693	0.60	1.693	0.60
10		1.623	0.58	1.623	0.58	1.627	0.58	1.632	0.58
100	cereals)	1.075	0.38	1.079	0.39	1.113	0.40	1.148	0.41
Background		0.224	0.08	0.224	0.08	0.224	0.08	0.224	0.08
1	Time	0.212	0.08	0.212	0.08	0.212	0.08	0.212	0.08
10	Tillie	0.130	0.05	0.131	0.05	0.132	0.05	0.133	0.05
100		0.012	0.00	0.013	0.00	0.015	0.01	0.018	0.01
Background		0.108	0.04	0.108	0.04	0.108	0.04	0.108	0.04
1	Melhus	0.107	0.04	0.107	0.04	0.107	0.04	0.108	0.04
10	(cereals)	0.097	0.03	0.099	0.04	0.102	0.04	0.105	0.04
100		0.046	0.02	0.056	0.02	0.074	0.03	0.091	0.03
Background		0.108	0.04	0.108	0.04	0.108	0.04	0.108	0.04
1	Melhus	0.107	0.04	0.107	0.04	0.107	0.04	0.107	0.04
10	(grass)	0.097	0.03	0.097	0.03	0.098	0.03	0.099	0.04
100		0.044	0.02	0.045	0.02	0.051	0.02	0.056	0.02

#### 9.1.3 Secondary poisoning of terrestrial mammals

The RCRs indicate a risk for secondary poisoning of terrestrial mammals at the present level of Cd content in soil at Stange (Table 9.1.3-1). The predicted concentrations show a decline with time, but the RCRs still remain above 1 after 100 years. For the Ås, Time, and Melhus scenarios, all RCRs are well below 1 and decrease with time, indicating no risk of secondary poisoning. The trends in relation to time and effect of Cd content in mineral P are the same as described in the previous section.

**Table 9.1.3-1.** Predicted environmental concentrations (PEC, mg Cd kg $^{-1}$  DW) and RCR for secondary poisoning of terrestrial mammals (PNEC = 0.9 mg Cd kg $^{-1}$ ), with application of mineral P fertilisers with the present, and alternative higher, content of Cd for 0-100 years. Bold data indicate RCR  $\geq$ 1.

		mg Cd kg <sup>-1</sup> Min.P fertiliser									
Year		Pres	ent	45	.8	91	6	13	7.4		
		PEC	RCR	PEC	RCR	PEC	RCR	PEC	RCR		
Background		0.282	0.31	0.282	0.31	0.282	0.31	0.282	0.31		
1	Ås	0.282	0.31	0.282	0.31	0.282	0.31	0.282	0.31		
10		0.280	0.31	0.281	0.31	0.283	0.31	0.286	0.32		
100		0.263	0.29	0.273	0.30	0.294	0.33	0.315	0.35		
Background		1.700	1.89	1.700	1.89	1.700	1.89	1.700	1.89		
1	Stange (cereals)	1.693	1.88	1.694	1.88	1.694	1.88	1.694	1.88		
10		1.636	1.82	1.637	1.82	1.640	1.82	1.643	1.83		
100		1.174	1.30	1.185	1.32	1.209	1.34	1.233	1.37		
Background	Stange (potato, carrot,	1.700	1.89	1.700	1.89	1.700	1.89	1.700	1.89		
1		1.692	1.88	1.692	1.88	1.693	1.88	1.693	1.88		
10		1.623	1.80	1.623	1.80	1.627	1.81	1.632	1.81		
100	cereals)	1.075	1.19	1.079	1.20	1.113	1.24	1.148	1.28		
Background		0.224	0.25	0.224	0.25	0.224	0.25	0.224	0.25		
1	Time	0.212	0.24	0.212	0.24	0.212	0.24	0.212	0.24		
10	rime	0.130	0.14	0.131	0.15	0.132	0.15	0.133	0.15		
100		0.012	0.01	0.013	0.01	0.015	0.02	0.018	0.02		
Background		0.108	0.12	0.108	0.12	0.108	0.12	0.108	0.12		
1	Melhus	0.107	0.12	0.107	0.12	0.107	0.12	0.108	0.12		
10	(cereals)	0.097	0.11	0.099	0.11	0.102	0.11	0.105	0.12		
100		0.046	0.05	0.056	0.06	0.074	0.08	0.091	0.10		
Background		0.108	0.12	0.108	0.12	0.108	0.12	0.108	0.12		
1	Melhus	0.107	0.12	0.107	0.12	0.107	0.12	0.107	0.12		
10	(grass)	0.097	0.11	0.097	0.11	0.098	0.11	0.099	0.11		
100		0.044	0.05	0.045	0.05	0.051	0.06	0.056	0.06		

# 9.2 Risk characterisation in aquatic organisms

The risk characterisation is based on the PEC<sub>soil</sub> as shown in Table 8.1-2, and the PNECs described in section 7.3.1.

Risk characterisations have been performed in aquatic organisms exposed to surface water and sediment and in terrestrial organisms exposed to soil, on the basis of predicted environmental concentrations (PEC) of Cd in the relevant compartments and the predicted no effect concentrations (PNEC) for each category of organisms. The so-called Risk Characterisation Ratio (RCR), defined as PEC/PNEC, is a quantitative expression of risk (ECHA, 2016). At RCRs higher than 1, unacceptable effects on organisms are likely to occur. The higher the ratio, the more likely it is that unacceptable effects may occur.

The estimated PECs in surface water, and the RCR based on the soil background concentrations, and with annual additions of Cd from fertilisers with different levels of Cd-content, are shown in Table 9.2-1.

The RCR is above 1 for all scenarios in Stange, indicating a potential risk for adverse effects on aquatic organisms at the present background concentration of Cd in soil. The predicted concentrations show a decline with time, but the RCRs are still above 1 after 100 years. As the annual input of Cd from fertilisers adds less than 1% to the background concentration in soil, even at the highest level of Cd in fertilisers (137.4 mg Cd kg<sup>-1</sup> P), the influence of fertiliser application on the PEC and RCR is insignificant after one year, but the significance increases with time. This is most obvious at sites with low background concentrations and high application rates of fertilisers, such as Melhus with cereal cultivation. For this scenario, the PEC and RCR after 100 years with the highest content of Cd in mineral P are 64% higher than with the present content of Cd in mineral P.

For Ås, the RCR is approximately 0.7, which means that no risk is predicted. The Cd concentrations increase slightly with time at the two highest levels of Cd in mineral P, and the RCR reaches 0.75 after 100 years with the highest ML for Cd in mineral P (137.4 mg Cd kg<sup>-1</sup>). This represents a 15% increase in RCR compared with the present level of Cd in mineral P applied for 100 years.

**Table 9.2-1.** Risk Characterisation Ratios (RCR) for aquatic organisms (PNEC =  $0.08 \ \mu g \ L^{-1}$ ), with application of mineral fertilisers with the present, and alternative higher, concentrations of Cd for 0-100 years. Bold data indicate RCR $\geq$ 1.

				mg C	Cd kg <sup>-1</sup> M	lin.P fertili	ser		
		Present		45.8		91.6		137.4	
Year		PECsw	RCR	PECsw	RCR	PECsw	RCR	PECsw	RCR
		μg Cd L <sup>-</sup>		μg Cd L <sup>-</sup>		μg Cd L <sup>-</sup>		μg Cd L <sup>-</sup>	
Background		0.055	0.69	0.055	0.69	0.055	0.69	0.055	0.69
1	Ås	0.055	0.69	0.055	0.69	0.055	0.69	0.055	0.69
10	AS	0.055	0.68	0.055	0.68	0.055	0.69	0.056	0.69
100		0.052	0.65	0.054	0.67	0.057	0.71	0.060	0.75
Background		0.244	3.05	0.244	3.05	0.244	3.05	0.244	3.05
1	Stange	0.243	3.04	0.243	3.04	0.243	3.04	0.243	3.04
10	(cereals)	0.235	2.94	0.235	2.94	0.236	2.95	0.236	2.95
100		0.172	2.15	0.173	2.17	0.177	2.21	0.180	2.25
Background	Stange (potato, carrot,	0.244	3.05	0.244	3.05	0.244	3.05	0.244	3.05
1		0.243	3.04	0.243	3.04	0.243	3.04	0.243	3.04
10		0.233	2.92	0.233	2.92	0.234	2.92	0.235	2.93
100	cereals)	0.158	1.98	0.159	1.98	0.164	2.04	0.168	2.10
Background		0.078	0.97	0.078	0.97	0.078	0.97	0.078	0.97
1	Time	0.074	0.93	0.074	0.93	0.074	0.93	0.074	0.93
10	Tillic	0.050	0.63	0.050	0.63	0.051	0.63	0.051	0.64
100		0.015	0.19	0.016	0.20	0.016	0.21	0.017	0.21
Background		0.017	0.21	0.017	0.21	0.017	0.21	0.017	0.21
1	Melhus	0.017	0.21	0.017	0.21	0.017	0.21	0.017	0.21
10	(cereals)	0.016	0.20	0.016	0.20	0.016	0.21	0.017	0.21
100		0.009	0.11	0.010	0.13	0.013	0.16	0.015	0.19
Background		0.017	0.21	0.017	0.21	0.017	0.21	0.017	0.21
1	Melhus	0.017	0.21	0.017	0.21	0.017	0.21	0.017	0.21
10	(grass)	0.016	0.20	0.016	0.20	0.016	0.20	0.016	0.20
100		0.009	0.11	0.009	0.11	0.010	0.12	0.010	0.13

# 9.3 Sediment-dwelling (benthic) organisms

The calculated PECs and RCRs are shown in Table 9.3-1.

**Table 9.3-1.** PECs and RCRs for sediment-dwelling organisms (PNEC =  $2.3 \text{ mg kg}^{-1}$ ), with application of mineral P fertilisers with the present, and alternative higher, content of Cd for 0-100 years. Bold data indicate RCR $\geq$ 1.

				mg Cd	kg <sup>-1</sup> Mi	in.P fertilise	r		
		Presei	nt	45.8		91.6		137.4	
Year		PECsediment	RCR	PECsediment	RCR	PECsediment	RCR	PECsediment	RCR
		mg Cd kg <sup>-1</sup>		mg Cd kg <sup>-1</sup>		mg Cd kg <sup>-1</sup>		mg Cd kg <sup>-1</sup>	
Background	Ås	0.52	0.23	0.52	0.23	0.52	0.23	0.52	0.23
1		0.52	0.23	0.52	0.23	0.52	0.23	0.52	0.23
10	Λ3	0.51	0.22	0.52	0.22	0.52	0.23	0.52	0.23
100		0.49	0.21	0.50	0.22	0.54	0.23	0.57	0.25
Background	Stange (cereals)	2.30	1.00	2.30	1.00	2.30	1.00	2.30	1.00
1		2.29	1.00	2.29	1.00	2.29	1.00	2.29	1.00
10		2.22	0.96	2.22	0.96	2.22	0.97	2.23	0.97
100		1.62	0.70	1.63	0.71	1.67	0.72	1.70	0.74
Background	Stange (potato, carrot,	2.30	1.00	2.30	1.00	2.30	1.00	2.30	1.00
1		2.29	1.00	2.29	1.00	2.29	1.00	2.29	1.00
10		2.20	0.96	2.20	0.96	2.21	0.96	2.21	0.96
100	cereals)	1.49	0.65	1.50	0.65	1.54	0.67	1.59	0.69
Background		0.73	0.32	0.73	0.32	0.73	0.32	0.73	0.32
1	Time	0.70	0.30	0.70	0.30	0.70	0.30	0.70	0.30
10	Timic	0.47	0.21	0.47	0.21	0.48	0.21	0.48	0.21
100		0.15	0.06	0.15	0.06	0.15	0.07	0.16	0.07
Background		0.16	0.07	0.16	0.07	0.16	0.07	0.16	0.07
1	Melhus	0.16	0.07	0.16	0.07	0.16	0.07	0.16	0.07
10	(cereals)	0.15	0.06	0.15	0.07	0.15	0.07	0.16	0.07
100		0.09	0.04	0.10	0.04	0.12	0.05	0.14	0.06
Background		0.16	0.07	0.16	0.07	0.16	0.07	0.16	0.07
1	Melhus	0.16	0.07	0.16	0.07	0.16	0.07	0.16	0.07
10	(grass)	0.15	0.06	0.15	0.06	0.15	0.06	0.15	0.07
100		0.08	0.04	0.08	0.04	0.09	0.04	0.10	0.04

The RCR at Stange is 1, indicating a risk to sediment-dwelling organisms with the background concentration of Cd in soil, and after one year with fertiliser application. But as the concentrations of Cd decrease with time, the RCRs are below 1 after 10 and 100 years, indicating no further risk of effects.

For all scenarios, the trends are the same as described for surface-water organisms. This is to be expected, as the concentrations of Cd in sediments are related to those in surface water.

## 9.4 Risk characterisation of farm animals

The model for Cd concentrations in feed and pasture plants, even at the highest level of mineral fertiliser tested for 100 years (worst case), found that the Cd level would decline for three of the municipality cases Time, Melhus and Stange. At Ås, it was estimated an accumulation of Cd in soil at 11.7% within a 100-year perspective.

In all selected geographical regions, including the alum shale area, the current Cd levels in feed and pasture plants, and thus the animal diet, are below a critical level of concern for consumers of animal products, and far below considered levels of animal health concern. By using mineral fertiliser, Cd concentrations in animal diets will mostly decrease and reduce the risk of Cd concentrations of concern in animal products, as well as the less-likely risk for adverse health effects in animals. However, livers and kidneys of older animals can enter the human food chain and Cd from these organs can potential be transferred to human via foods of animal origin.

Exposure of animals to Cd levels via their drinking water is usually far below that from their feed and from pasture plants, and is not expected to contribute significantly to their total exposure.

## 9.5 Risk characterisation for humans

The TWI for humans is based on long-term accumulation of Cd in kidneys, leading to toxic concentrations after many years. The risk for acute effects due to a slight increase in Cd concentrations in leafy vegetables, currently contributing to 3.9% of the dietary exposure, is negligible. The TWI is based on early markers of kidney failure, and long-term exposure below this TWI is considered safe, even if the TWI is exceeded for a limited period. This risk assessment is therefore based on long-term changes in Cd concentrations in food plants. The current mean dietary exposure of Cd is estimated to exceed the TWI for the youngest age groups, and for most age groups for the high consumers (see Table 7.3.3.2-1.). Even adults have a Cd exposure close to the TWI. A reduction in Cd exposure is therefore desirable, as this will reduce the risk in the population from toxic effects due to Cd. A reduction in Cd concentrations in the main dietary sources, such as cereals, potatoes, and root vegetables, will reduce dietary exposure and the proportion of the population with a dietary exposure above the TWI will decrease. Modelling indicates that the differences between the selected Cd concentrations are moderate and will not have any significant impact on total dietary exposure. Independent of the modelled scenarios, the estimates

indicate that Cd concentrations in crop plants will decrease, thereby reduce exposure and the health risk for the population.

The predicted levels of Cd in crop plants from the Stange region are high (see Table 8.3-1a) However, it is unlikely that any consumer will purchase plants only cultivated in soil with naturally high Cd concentrations. Nevertheless, consumers of large quantities of home-grown vegetables from this area, and consumers purchasing directly from a local farm, are likely to exceed the TWI, regardless of use of fertilisers and cultivation practices.

VKM has evaluated the risk to children eating soil, taking into account that this exposure route usually only occurs in a limited age range. Given the low estimated exposure compared with intake from the diet, this exposure route is considered to be of low risk.

### 10 Uncertainties

# 10.1 Uncertainty assessment and intrinsic uncertainty of the assessment method

The risk assessment procedure involves three stages; Hazard assessment, Exposure assessment and Risk characterisation and there are uncertainty in each of them (ECHA, 2012).

For proper quantification of the uncertainty of the estimated outcome, the distribution, variance, and, thus, uncertainty, of all sensitive input parameters needs to be assessed (Trapp and Matthies, 1998). This is beyond the scope and time limit of the current assessment. However, some principle uncertainties of the underlying assessment method can be identified, as listed below:

- Analysis of Cd in mineral P fertilisers, and other input sources, background concentrations.
- Fate:
  - The fate model of Cd applied in this risk assessment is based on a box model
    of the top soil with regionally averaged input data. This is a common and
    convenient approach, and mathematically easy to handle. It also makes
    calculations easy because the true (and well-known) spatial heterogeneities of
    soil and environment are combined into a single dataset.
  - The input data used for the calculation of exposure and risk were considered to be as precise and as close to the true situation as possible, but were averaged from many single measurements. The calculation can therefore also only give an average result, representing a typical, common scenario. Extreme, rare, and thus unlikely, conditions were not considered. This is the assessment that is usually required and this approach is also common practice in other risk assessments (EU 2007, Smolders 2013, SCHANT, 2015; SCHER, 2015).
  - There are uncertainties connected to choosing representative parameters and factors (e.g., soil quality and climate related parameters), for instance, infiltration rate in the different regions are based on assumptions.
- Risk evaluation in farm animals and humans:
  - The Cd concentrations in the various feed plants are based on mean values and the presentation does not consider the variation of cadmium in the feed plants. Thus, some animals would be exposed to higher and other to lower cadmium concentrations. However, mean yearly concentrations are assumed

- to represent the realistic average total levels in the situation of repeated application of the fertiliser for many years.
- In the hazard characterisation, there is somewhat uncertainty of the tolerable daily intake of cadmium in the different animal species.
- There are uncertainties related to human dietary exposure. There is insufficient Norwegian data on Cd in all food items to estimate exposure in the Norwegian population, and therefore exposures are based on equivalent estimates from EU. Furthermore, there are uncertainties related to the use of standard body weights used by VKM in its previous assessment (VKM 2015). The proportion of the total crop on the market that is produced under the different modelled scenarios is unknown. It should be noted that dietary habits are changing continuously, and are likely to change significantly within a time period of 100 years.
- There are also uncertainties related to the TWI, as discussed in EFSA (2012).
   These uncertainties remain valid.
- Human toxicity thresholds for chronic exposure are commonly derived for lifelong exposure. However, nowadays, consumers in most, if not all, cases obtain their food from a variety of sources, local and global. It can therefore be expected that during the life-span, extreme events, with high short-term exposure, are balanced with longer periods of low exposure.

#### Environmental risk assessment

- In the hazard assessment, PNECs for various groups of exposed organisms are derived, based on toxicity tests in which a single species is usually exposed to a series of concentrations or doses of a substance, and acute or chronic toxic effects recorded. Test endpoints are typically LC50, EC50 for acute responses, and EC10 or NOEC for chronic responses. After compilation of all relevant endpoints, the PNEC is calculated initially applying an assessment factor on the lowest ("most sensitive") endpoint. Which assessment factor to use is dependent on the quality and quantity of the available test data. The assessment factors are used to address the uncertainty involved in extrapolating from a limited number of species, and endpoints tested in the laboratory, to the field situation where "all" species" must be considered regarding any adverse effect posed by exposed to the substance.
- For Cd, a wealth of data exists on the toxic effects of Cd to terrestrial and aquatic organisms. Relevant data for derivation of PNECs have been compiled in the European risk assessment report (EC, 2007). The high amounts of data and species represented allow the use of statistical approaches to calculate PNECs for aquatic and terrestrial organisms. The remaining uncertainty in the hazard assessment is therefore considered as low.

- o In the exposure assessment, models have been used to calculate exposure concentrations in soil (PEC<sub>soil</sub>)and surface water (PEC<sub>sw</sub>). Uncertainty and critical parameters in the model used for calculation of PEC<sub>soil</sub> have been discussed under "Fate" above. Since the calculation of PEC<sub>sw</sub> uses output from the fate model as a basis for further calculation of exposure concentration in surface-water recipients, the same considerations of uncertainty apply here. Additional critical parameters for calculation of PEC<sub>sw</sub> are the concentrations of suspended solids in the receiving water (SUSP<sub>WATER</sub>), the partitioning constant for suspended solids and water (Kp<sub>susp</sub>), and the dilution factor.
- SUSP<sub>WATER</sub> has been set as 15 mg L<sup>-1</sup>, which is the recommended default value in TGD (ECB, 2003). Suspended solids have the effect of reducing the PEC<sub>SW</sub>, as demonstrated in the sensitivity tests below. The variation in content of suspended solids in surface water in Norway is very high, and lower values than 15 mg L<sup>-1</sup> occur frequently. However, small streams in agricultural areas will often have a much higher content of suspended solids, especially during episodes when drainage water from agricultural soils are discharged; therefore, the use of 15 mg L<sup>-1</sup> is considered sufficiently conservative.
- Kp<sub>susp</sub> has been adopted from the European risk assessment report on Cd (EC, 2007), and is an average value for European freshwaters. Factors that influence the actual Kp-value are pH, total metal concentration, water hardness, and the nature and concentrations of complexing agents. No analyses of these parameters in relevant Norwegian surface waters are available, which indicates these parameters as a possible uncertainty factors. The effect of Kp<sub>susp</sub> is to reduce the PEC<sub>sw</sub>, as shown in the sensitivity test below.
- The dilution factor has been set at 10, which is the recommended default value in TGD (ECB, 2003). Considerable local variations in dilutions of agricultural drainage waters can be expected. Less than 10 times dilutions occur in open ditches adjacent to cultivated fields. For the scenario that has been used for the risk assessment (i.e. small, permanent streams or rivers running through agricultural areas, but with a significant part of the drainage area that is not cultivated), a dilution factor of 10 is a reasonable assumption. The few field measurements of Cd that are available from such streams in the areas that have been selected for this assessment indicate that the PEC<sub>SW</sub> has not been underestimated, as discussed in 7.4.2.
- The risk characterisation provided Risk Characterisation Ratios (RCR) well above 1, indicating risks for adverse effects on aquatic organisms and terrestrial organisms by secondary poisoning, in the alum shale area in Stange. The reason for this is the high content of geogenic Cd in the soil. In this assessment, there has been no distinction between anthropogenic and geogenic Cd in terms of mobility and bioavailability. As Cd from alum shale is

less mobile and available for uptake than Cd from anthropogenic sources such as fertilisers, mobility/bioavailability are important uncertainty factors in this risk characterisation. The direction of this uncertainty is overestimation of risk.

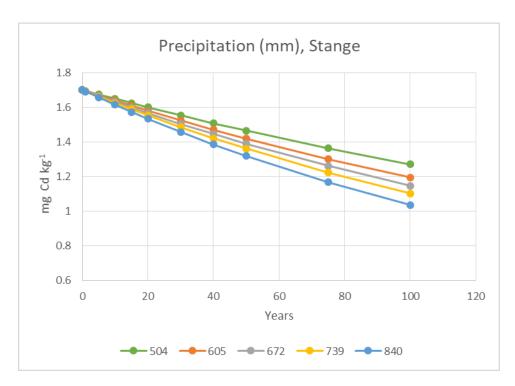
### 10.2 Sensitivity

### 10.2.1 Sensitivity analyses of PEC<sub>soil</sub> calculations

Sensitivity analyses were performed to evaluate the effects of the following parameters on the  $PEC_{soil}$  up to 100 years at Ås and Stange: precipitation, SOM, Kd, and pH. The simulations are based on the scenarios with maximum ML for Cd in mineral P (137.4 mg Cd kg<sup>-1</sup> P) and maximum application of sludge (Ås).

For pH, a range between 4.6 and 6.6 ( $\pm$  1 pH unit from the value used in the PEC<sub>soil</sub> calculations) was selected. For the other parameters, + and - 10 and 25% were selected as variations in the sensitivity analyses. In the figures 9-1 - 9.4, grey markers and lines are used for the parameter values used in the PEC<sub>soil</sub> calculations (section 7.4.1).

<u>Precipitation:</u> The Cd loss rate increases with precipitation. At Stange, where accumulation of Cd was predicted with the value used in calculation (672 mm yr<sup>-1</sup>), this was almost eliminated with a 25% increase in precipitation (to 840 mm). For Stange, the concentrations of Cd decreased with time at all precipitation levels tested (Fig. 10.2.1-1).



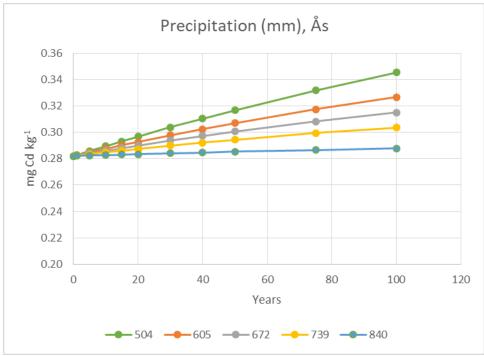
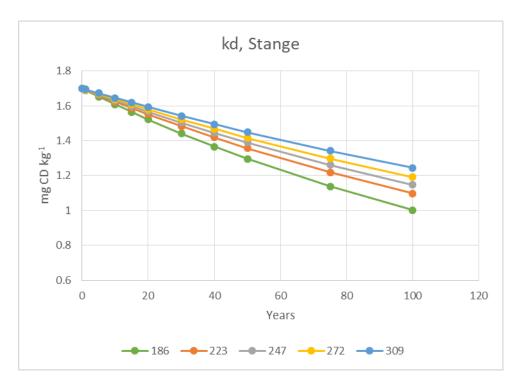


Fig. 10.2.1-1. Sensitivity test precipitation at: a) Stange, and b) Ås.

 $\underline{\text{Kd:}}$  Cd loss rate decreases with increasing Kd. At Ås, Cd accumulation occurs at Kd values above 163, where the Cd concentration is at steady state at 0.28 mg Cd kg<sup>-1</sup> (Fig. 10.2.1-2).



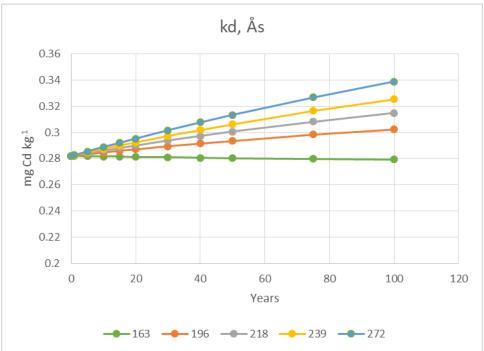
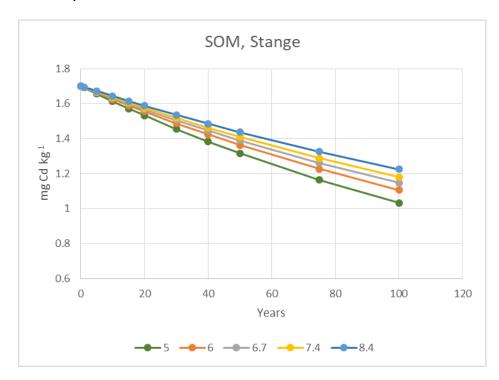


Fig. 10.2.1-2. Sensitivity tests of Kd at: a) Stange, and b) Ås. (Note difference in scales).

<u>SOM:</u> Cd loss rate decreases with increasing SOM concentration. Reducing the SOM concentration by 25% to 4.3 mg  $L^{-1}$  gives almost steady state concentration of Cd (Fig. 10.2.1-3).



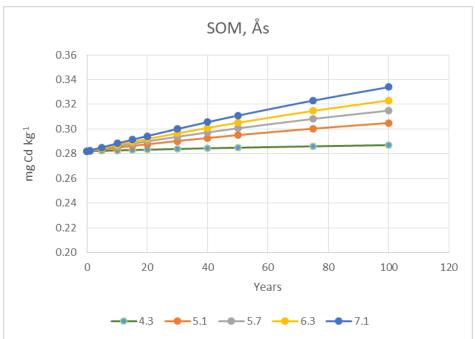
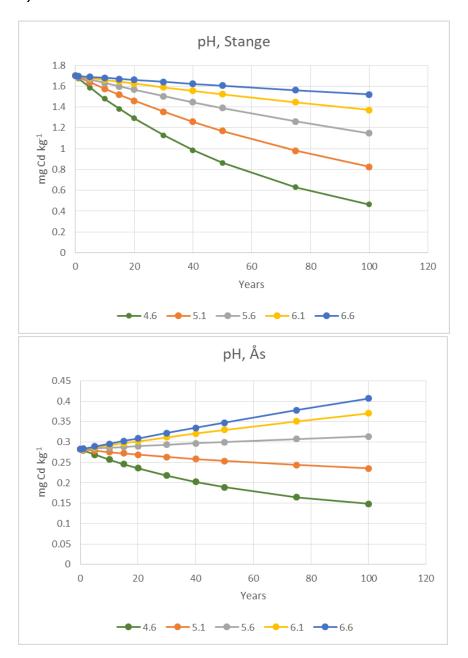


Fig. 10.2.1-3. Sensitivity tests of SOM at: a) Stange, and b) Ås. (Note difference in scales).

<u>pH:</u> Cd loss rate decreases with increasing pH. Reducing the  $pH(CaCl_2)$  by 0.5 units to 5.1 reversed the accumulation to a negative trend of Cd in soil at Ås. At Stange, the Cd concentration declines at all pH-values tested. Approximately 80% of the analysed soil samples from Ås and Stange fall between the red and yellow lines in the graphs (Fig. 10.2.1-4).

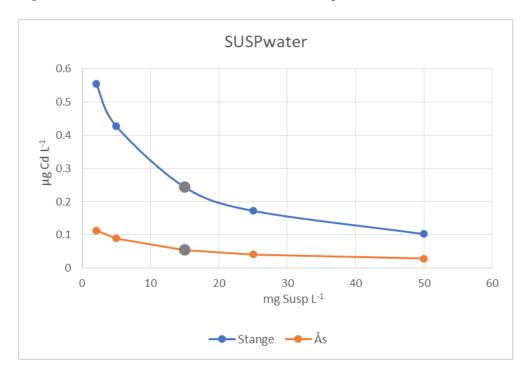


**Fig. 10.2.1-4.** Sensitivity tests of pH (CaCl<sub>2</sub>) at: a) Stange, and b) Ås. (Note difference in scales).

### **10.2.2** Sensitivity testing of PEC<sub>sw</sub> calculations

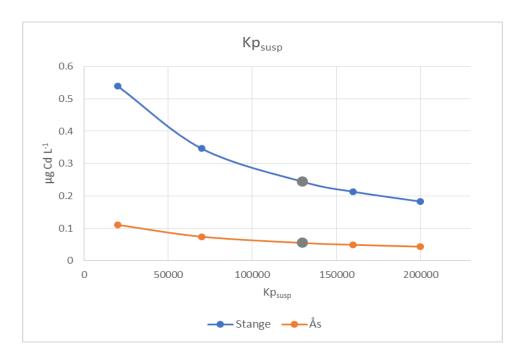
Predicted concentrations of Cd in Surface Waters (PEC<sub>SW</sub>) are calculated on the basis of the concentration of Cd in drainage water using Equation 9, (See section 8.2.2). Sensitivity tests have been made on the parameters SUSP<sub>water</sub>, kp<sub>susp</sub>, and dilution rate. The tests have been made on the present background scenarios at Stange and Ås. In the figures 10.2.2-1 to 10.2.2-3, the parameter values used for calculation of PEC<sub>SW</sub> in Section 8.2.2 are indicated by grey markers.

SUSP<sub>WATER</sub>: A reduction of SUSP<sub>WATER</sub> from the default value (15 mg L<sup>-1</sup>, ECB 2003) to 5 mg L<sup>-1</sup>, gives a 75% increase in PEC<sub>SW</sub> at Stange and 63% at Ås. An increase of SUSP<sub>WATER</sub> to 50 mg L<sup>-1</sup> results in 58% decrease in PEC<sub>SW</sub> at Stange, and 49% at Ås.



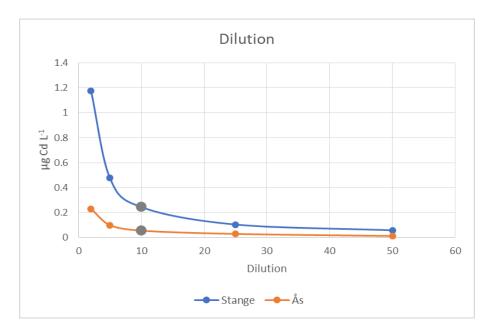
**Fig. 10.2.2-1.** Effect of parameter SUSP<sub>WATER</sub> on the PEC<sub>SW</sub> at Stange and Ås, based on the present background concentration of Cd in soil.

 $\underline{\mathsf{KP}_{\mathsf{susp}}}$ : In the calculations of  $\mathsf{PEC}_{\mathsf{SW}}$ ,  $\mathsf{Kp}_{\mathsf{susp}}$  was set at 130 000 (EC, 2007). A lower value gives higher  $\mathsf{PEC}_{\mathsf{SW}}$ . A reduction of  $\mathsf{Kp}_{\mathsf{susp}}$  to 70 000 results in a 41% increase of  $\mathsf{PEC}_{\mathsf{SW}}$  at Stange, and 35% at Ås.



**Fig. 10.2.2-2.** Effect of parameter  $Kp_{susp}$  on the  $PEC_{SW}$  at Stange and Ås, based on the present background concentration of Cd in soil.

<u>Dilution rate</u>: The dilution rate used to calculate  $PEC_{SW}$  in Section 7.4.2 was 10, which is the recommended default dilution in TGD (ECB, 2003). A 50% reduction in the dilution rate to 5, gives a 42% increase in  $PEC_{SW}$  at Stange, and 35% at Ås.



**Fig. 10.2.2-3.** Effect of parameter dilution rate, on the PEC $_{SW}$  at Stange and Ås, based on the present background concentration of Cd in soil.

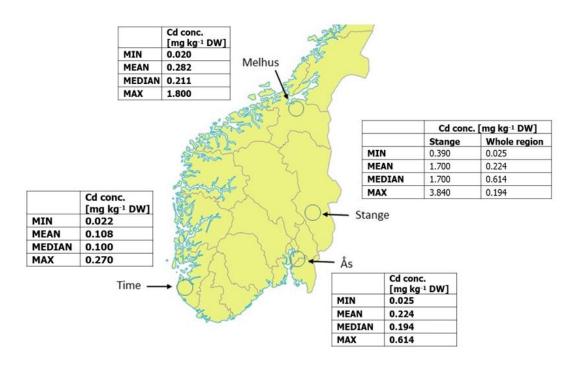
# 11 Conclusions and answers to the terms of reference

In this risk assessment, four different municipality case areas covering major agricultural practices in Norway were chosen: 1) Southeastern Norway, with Ås municipality as a case area; 2) Hedmark region, with Stange municipality as a case area for alum shale; 3) Southwestern Norway, with Time municipality as a case area; and 4) Trøndelag (Mid-Norway), with Melhus municipality as a case area.

## 1. What do we know about the levels of cadmium in agricultural soils in Norway today?

In general, the data available for Norwegian agricultural soil indicate low natural geogenic Cd concentrations (overall median < 0.2 mg kg<sup>-1</sup>). However, areas with alum shales can show clearly higher values (Fig. 11-1). A total of 78 analyses for Cd in agricultural soil from 26 different municipalities in Trøndelag were included here, and the median value was 0.1 mg Cd kg<sup>-1</sup>. In Southwestern Norway (Rogaland and Vest-Agder County), a higher median Cd concentration was calculated (0.19 mg Cd kg<sup>-1</sup>), however, only 43 samples from 17 municipalities were included. Southeastern Norway, covering the counties Østfold, Akershus, and Vestfold, is the region with the second highest Cd levels in agricultural soil, with a median of 0.21 mg Cd kg<sup>-1</sup> based on 108 samples from 27 municipalities. Out of the 108 samples, 47 were from Sandefjord, which thus dominates the data collection. However, Sandefjord shows a similar concentration range of Cd as the whole region (Appendix I). Hedmark is the region showing the highest Cd concentrations in arable land due to areas with alum shales (e.g., in Stange municipality). Data from Stange municipality indicate a maximum Cd concentration in agricultural soil of 3.8 mg Cd kg<sup>-1</sup> and a median of 1.7 mg Cd kg<sup>-1</sup>. It should be noted that samples from Stange were collected in connection with studies of agricultural soils with alum shale 'problems'. Hedmark, in general, does not have higher concentrations of Cd than the other regions.

The median and average Cd concentrations in soil in Ås (0.2 and 0.28 mg kg<sup>-1</sup> DW) were higher than in Time (0.19 and 0.22 mg kg<sup>-1</sup> DW) and Melhus (0.1 and 0.11 mg kg<sup>-1</sup> DW). Six and Smolders (Six and Smolders, 2014) reported that around 0.3 mg Cd kg<sup>-1</sup>, which is slightly higher than these values, was realistic for use as a background concentration in evaluating Cd in P fertilisers.



**Fig. 11-1.** Illustration of present (background) concentrations of Cd in the regions included in this risk assessment.

2. Describe the fate (mobility) of cadmium in agricultural soil and in the local environment, after the application of cadmium-containing mineral phosphorus fertilisers to agricultural land.

**Fate and mobility of Cd — influencing parameters and long-term change in Cd concentration in soil:** The long-term changes in Cd concentrations in soil depend highly on its sorption properties to soil. Sorption is described by the distribution coefficient, Kd, which indicates the binding/mobility properties of Cd and is expressed as Cd concentration in soil divided by concentration in pore water (presented as L kg<sup>-1</sup>). A high Kd value indicates higher binding to soil and organic matter than lower Kd values, and, thus, lower leaching, but higher accumulation in soil, and higher predicted uptake and transfer to plants. The Kd value, and the parameters influencing the Kd value, are important for leaching of Cd from soil to nearby recipients (groundwater and surface water) and for bioavailability and transfer to plants and other organisms.

The most important soil parameters influencing sorption processes and the long-term change of Cd concentrations in soil, are soil pH and soil organic matter (SOM) (see section 10, Six and Smolders, 2014). The initial Cd concentration in soil is also an important parameter, but of lesser importance than pH (Six and Smolders, 2014). Clay content and cation exchange capacity are other parameters that are known to influence sorption of Cd (Six and Smolders, 2014). In addition to soil parameters, loss of Cd from soil is also influenced by the climate, particularly precipitation and precipitation infiltration (section 10). There are high uncertainties connected with selecting infiltration rate in the different regions. Annual precipitation in different regions have been recorded. Increased precipitation and more intense rain events are predicted. Also regional differences is predicted; e.g. towards 2100 there is expected to be an increase of about 10-15% in annual precipitation in southwestern and mid Norway, and 0-15% in southeastern Norway.

Soil quality properties and climate data were selected from the NIBIO Soil database and different monitoring programmes, and some are based on values used in a previous health and environmental risk assessment performed by VKM (VKM, 2014). For most parameters, mean values were selected for modelling. One exception was made, for predicting the contribution of Cd in sewage sludge. Here, both the realistic worst-case scenario with maximum allowed application according to the regulation (as a conservative approach) and a scenario based on reported application rate of sewage sludge by Statistics Norway (SSB) were investigated. Application of sewage sludge was only accounted for in the two municipality cases in southeastern Norway, Ås and Stange (solely cereal production). Cereals are the dominant crops in these regions. However, potato and carrot also have high cultivation yields in Stange, and, as root vegetables have higher P requirements and use of mineral P fertilisers, a second risk evaluation with the crop rotation potato, carrot, and cereals was included. Stange municipality represents agricultural cultivation on alum shale with a high natural (geogenic) Cd content, but it is important to note that alum shale and high geogenic Cd in background soil is not representative for the whole Hedmark County, neither all agricultural land in Stange. Grass is the main crop in Time, and in Melhus both grass and cereals are important crops and included as crop rotations.

Predicted Cd contributions via worst-case scenario application of sewage sludge was 2.4 g Cd ha<sup>-1</sup> yr<sup>-1</sup>, and very much higher than the average used sewage sludge ( $\leq 0.03$  g Cd ha<sup>-1</sup> yr<sup>-1</sup>), based on data from SSB (Fig. 11-2a). The predicted Cd contribution via liming products was very low, being  $\leq 0.013$  g Cd ha<sup>-1</sup> yr<sup>-1</sup> in all regions. The contribution via atmospheric deposition is 0.34 g Cd ha<sup>-1</sup> yr<sup>-1</sup> (same in all scenarios), and manure (accounted for at Melhus and Time municipalities) contributes in the range of 0.24-0.54 g Cd ha<sup>-1</sup> yr<sup>-1</sup>. The contribution of Cd via mineral P fertiliser depends on the type of crop (P requirement) and

<sup>&</sup>lt;sup>2</sup> https://www.environment.no/

the type of mineral fertiliser applied. The modelling was based on the reported Cd content of mineral P fertilisers in Norway and current fertiliser practices for the different crop rotation for the given municipalities. The Cd contributions with mineral P fertiliser (based on the most common used NPK fertilisers in these scenarios , range 25-50 mg Cd kg<sup>-1</sup> P, 2015 data) and the current agricultural practices were predicted to be about 0.14-0.17 g Cd ha<sup>-1</sup> yr<sup>-1</sup> for grass production at Time and Melhus, around 0.32-0.42 g Cd ha<sup>-1</sup> yr<sup>-1</sup> for cereal production at Melhus, Ås, and Stange, and highest at Stange with the crop rotation potato, carrot, and cereals with 0.99 g Cd ha<sup>-1</sup> yr<sup>-1</sup> (Fig. 11-2a). The predicted Cd contributions with increasing maximum limit (ML) in mineral P fertiliser were around 0.7 g Cd ha<sup>-1</sup> yr<sup>-1</sup> for grass production, around 1.7-2.3 g Cd ha<sup>-1</sup> yr<sup>-1</sup> for cereal production, and 3.3 g Cd ha<sup>-1</sup> yr<sup>-1</sup> for the potato, carrot, and cereals crop rotation.

Time, in southwestern Norway, is the case municipality with the highest precipitation (1464 mm yr<sup>-1</sup>) and infiltration rate (0.7, given as infiltrated fraction of precipitation), but also the lowest mean soil pH (5.3 in CaCl<sub>2</sub>) and SOM content (4.1%). These soil qualities, together with a high precipitation rate, result in the highest predicted Cd removal kinetic, both via leaching and via crop harvesting (crop offtake). The low pH and SOM, both resulted in predicted low Kd (115.9 L kg<sup>-1</sup> compared with Kd 217.6 at Ås, 247.4 at Stange, and 257.8 at Melhus). Independent of maximum limit (ML) of Cd in mineral P fertiliser, the Cd input rate was considerably below the Cd removal rate, and the Cd reduction in soil from a 100-year perspective was predicted to be around 90-95%. Since the use of mineral P is low in this region, the effect of change in ML of Cd, is low.

The municipality case of Melhus, in mid Norway, also has a high precipitation (1236 mm yr<sup>-1</sup>) and infiltration rate (fraction 0.7), but with a higher mean pH and SOM content than Time (5.7 and 6.3%, respectively). Predicted Cd removal kinetic was high compared with Cd input. This was also the case for the highest evaluated ML at 137.5 mg Cd kg<sup>-1</sup> P, and a long-term decline in soil concentration was predicted. The reduction in soil concentration of Cd from a 100-year perspective was in the range of 30 to 60% (cereal and grass production).

Precipitation and infiltration rate are lower in the southeastern region, and 672 mm precipitation yr<sup>-1</sup> and infiltration fraction 0.4 were chosen for the municipalities Stange and Ås. The mean pH is the same for these municipalities (5.6), but the mean SOM content is lower at Ås than Stange (5.7% versus 6.7%). For both crop rotations evaluated at Stange, a long-term net decline in soil Cd concentration was predicted. This trend was independent of the realistic worst-case sewage-sludge application approach - defined as the maximum allowed application rate of class I sewage sludge (40 tonnes ha<sup>-1</sup> 10 yr<sup>-1</sup> with Cd concentration < 0.8 mg Cd kg<sup>-1</sup> DW) - or an average application of sewage sludge in the municipality based on data from SSB, and with use of ML 137.5 mg Cd kg<sup>-1</sup> P. In a 100-year perspective, a 27 to 37% reduction in the soil concentration of Cd was estimated.

At Ås, the situation was different. Using the chosen soil quality parameters, annual precipitation and infiltration rates, and a realistic worst-case sewage sludge application, ML values of 91.6 and 137.4 mg Cd kg<sup>-1</sup> P, an accumulation of Cd in soil was estimated in the long-term perspective. The contribution of Cd via sewage sludge was the key factor in this case. Under the same conditions, but applying an average amount of sewage sludge for the municipality (based on data from SSB), the total input of Cd to soil decreased and changed the Cd mass balance in soil. This resulted in a net Cd removal and predicted reduction of Cd soil concentration in the long-term. The same annual precipitation and infiltration rates were chosen for Ås and Stange. However, more detailed evaluation of recorded precipitation data shows that this is somewhat higher in Ås than Stange.

We have used average Cd concentrations in manure, sewage sludge, liming products, and atmospheric contributions to estimate Cd application inputs to arable soil. As these values are not conservative, to account for this and to prevent the Cd application to soil in combination with sewage sludge being underestimated, the maximum allowed sewage sludge (according to the regulation) is included as a realistic worst-case scenario.

Based on the selection of equations, input parameters etc. in this risk assessment, a relatively high loss of Cd via leaching was predicted, particularly for Time and Stange. Due to the high annual precipitation and the assumed high infiltration rate being among the driving factors, leaching might be overestimated. For instance, during periods with frozen ground, and in rain events in which the contact time might be too short to establish Cd equilibrium between the soil and water phases, the Cd concentrations in solution will be lower than predicted based on Kd.

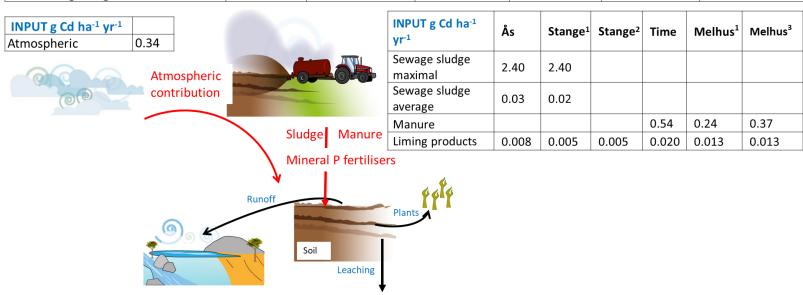
Several previous risk assessments have used Kd algorithms. However, these are now assumed to be too high, and might have resulted in overestimates of Cd concentrations in soil and underestimates of Cd concentrations in soil solutions and leachates from soil. An overestimates of soil concentrations will also result in overestimating the exposure of farm animals, and thereby humans, via their diet (e.g., grass, cereals, leafy and root vegetables). On the other hand, it will also underestimate the transfer from soil to surface water and groundwater.

**Fate and mobility of Cd – removal via harvesting of crops:** Removal of Cd from soil via harvested plants (also termed output via crop offtake), depends on the soil concentration and the crop yield for the given plant. However, in comparison with leaching and runoff, crop offtake plays only a minor role in the total removal of Cd from soil. The highest removal via this mechanism was predicted at Stange, with the crop rotation potatoes-carrots-cereals, accounting for a removal rate of 2.9 g Cd ha<sup>-1</sup> yr<sup>-1</sup> (Fig. 11-2b). A higher Cd removal via grass harvesting at Time compared with Melhus (1.2 versus 0.2 g Cd ha<sup>-1</sup> yr<sup>-1</sup>), is primarily due to the intensive grass production and higher annual yield in Time. Cd removal via harvest of cereals at Ås and Melhus were calculated to be 0.2 and 0.06 g Cd ha<sup>-1</sup> yr<sup>-1</sup>,



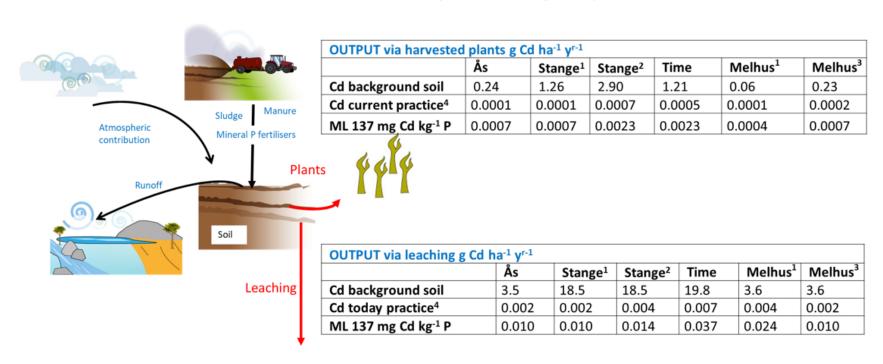
### Estimated input of Cd to agricultural soil

INPUT via Mineral P fertiliser g Cd ha <sup>-1</sup> y <sup>r-1</sup>						
	Ås	Stange <sup>1</sup>	Stange <sup>2</sup>	Time	Melhus <sup>1</sup>	Melhus <sup>3</sup>
Current practice (25-50 mg Cd kg P <sup>-1</sup> )	0.37	0.42	0.99	0.14	0.32	0.17
ML 43 mg Cd kg P <sup>-1</sup>	0.68	0.76	1.11	0.23	0.58	0.24
ML 91 mg Cd kg P <sup>-1</sup>	1.36	1.53	2.23	0.46	1.15	0.49
ML 137 mg Cd kg P <sup>-1</sup>	2.04	2.29	3.34	0.69	1.73	0.73



**Fig. 11-2a.** Illustration of estimated input of Cd via mineral P fertilisers, atmospheric contribution, sewage sludge (maximum allowed and average based on statistics), manure, and liming products, shown as g Cd ha<sup>-1</sup> yr<sup>-1</sup>. <sup>1</sup>Crop rotation cereals, <sup>2</sup>Crop rotation potato-carrot-cereals, <sup>3</sup>Crop rotation grass.

### Estimated removal via leaching and harvesting of crops



**Fig. 11-2b.** Illustration of predicted removal of Cd via harvesting (Eq. 5) and water transport via leaching (Eq. 2) from background soil, Cd added in mineral P fertilisers with current fertiliser practise and Cd content, and with highest ML 137 mg Cd kg P<sup>-1</sup>. <sup>1</sup>Crop rotation cereals, <sup>2</sup>Crop rotation potato-carrot-cereals, <sup>3</sup>Crop rotation grass, <sup>4</sup>In this risk assessment used 25-50 mg Cd kg<sup>-1</sup> P. Removal is presented as g Cd ha<sup>-1</sup> yr<sup>-1</sup>.

**Predicted transfer of Cd to surface water:** The excess precipitation (precipitation minus evaporation) from agricultural areas may reach surface waters through surface runoff and drainage. Transfer of Cd to surface water has been calculated with the assumption that all excess precipitation will drain to a surface water recipient, and will contain Cd extracted from the soil as described by the Kd. The resulting concentration of Cd in receiving surface waters (PEC<sub>SW</sub>) has been predicted for a standard scenario in which drainage water is diluted by a factor of 10 in a stream, according to recommended procedures for local assessments in technical guidance documents (ECB, 2003; ECHA, 2016). The predicted concentration in sediments (PEC<sub>sediment</sub>) has been calculated using a sediment/water partitioning constant (Kp), as described in the European Risk Assessment of Cd (EC, 2008).

Based on the present background concentrations in soil, the PEC<sub>SW</sub> surface water is 0.055  $\mu$ g L<sup>-1</sup> in Ås, 0.244  $\mu$ g L<sup>-1</sup> in Stange, 0.078  $\mu$ g L<sup>-1</sup> in Time, and 0.017  $\mu$ g L<sup>-1</sup> in Melhus. The PECs are supported by the few available data of Cd in streams in the same areas.

The PEC<sub>sediment</sub> is 0.52 mg kg<sup>-1</sup> in Ås, 2.30 mg kg<sup>-1</sup> in Stange, 0.73 mg kg<sup>-1</sup> in Time, and 0.16 mg kg<sup>-1</sup> in Melhus.

**Predicted transfer of Cd to forage and food:** Uptake and transfer of Cd into plants depends on the plant species, the soil concentrations of Cd, and the same soil properties as already mentioned, such as pH and organic matter. The Cd concentration in crops increases with increasing soil Cd concentration, given than soil parameters, precipitation, and other factors remain constant. Uptake in plants is expressed as a transfer factor (TF) (the concentration of Cd in plant tissue divided by the Cd concentration in soil). In this risk assessment, the TF for the most relevant plant species (cereals, potatoes, other vegetables (e.g., carrots), leafy vegetables, grass) were evaluated before TFs were chosen. The TFs, based on dry weight, varied from 0.165 (cereals) to 0.36 (grass and leafy vegetables). The chosen TFs for potatoes and carrots were 0.27 and 0.25, respectively. Predicted Cd concentrations in crops grown in different regions varied highly. However, they were clearly highest in plants produced on alum shale in Stange. For evaluating risk to humans, the exposure concentration is determined on a fresh weight basis. The TFs expressed in fresh weight and accounting for water content are: cereals 0.14, leafy vegetables 0.108, potatoes 0.081, carrots 0.075, and grass 0.072.

The predicted Cd concentrations in potatoes, carrots, and cereals grown in alum shale soil in Stange were 137.7, 127.5, and 238.4  $\mu$ g Cd kg<sup>-1</sup> FW, respectively, which are all above the ML values for Cd in the respective crops. With the exception of those crops grown on alum shale in scenario Stange, none of the predicted plant concentrations were above the ML values for Cd in crops.

Other crops cultivated on alum shale areas in Hedmark region have recently been analysed for Cd (Salbu et al., 2013); reported concentrations have been close to, but not above, the

limit values for Cd concentrations in crops, which is 0.1 mg kg<sup>-1</sup> FW for potato (peeled), carrots, barley, and oats, 0.2 mg kg<sup>-1</sup> FW for wheat, and 1 mg kg<sup>-1</sup> FW for grass (Table 8.3-3).

In this risk assessment, several assumptions have been made and input parameters have been selected, all of which have an associated uncertainty (see section 10). However, it is important to note that geogenic Cd has been shown to be less bioavailable than anthropogenically introduced Cd to soil. Anthropogenically contaminated soils showed more than 3 times higher availability than geogenic alum shale, even when geogenic-Cd soil contained nearly 20 times the amount of Cd than soil in which Cd addition has been anthropogenic (Kashem and Singh, 2002).

**Predicted effect of ML values for Cd in mineral P fertilisers on environmental fate of Cd:** The effect of application of mineral P fertilisers to agricultural soil over time has been predicted by comparing Cd input to agricultural soil with loss of Cd from the soil, both given as g Cd ha<sup>-1</sup> yr<sup>-1</sup>. It was predicted a decline of the Cd concentration in soil over time. This reduction is due to a higher loss rate than input rate of Cd to agricultural soil; this including the sum of fertilisers (both mineral and organic fertilisers and liming products) and atmospheric contribution.

Compared to a scenario with use of mineral P fertilisers containing the Cd concentration measured in most commonly used fertiliser types (NPK 22-3-10, 25-2-6, 12-4-18, and with a Cd concentration in the range of 25-50 mg Cd kg $^{-1}$  P, 2015 data), increasing ML level of Cd in mineral P fertilisers to 137.4 mg Cd kg $^{-1}$  P (60 mg Cd kg $^{-1}$  P $_2$ O $_5$ ), resulted in an estimated clear decline over time at Stange, Time and Ås.

However, it might lead to an accumulation of Cd where input Cd application rate is higher than removal rate from soil. In this risk assessment, one of the locations, Ås, indicate that an accumulation of Cd in soil over time might occur, but only if maximum allowed application of sewage sludge according to the present regulation is used (a realistic worst-case scenario). A ML level 91.6 mg Cd kg<sup>-1</sup> P, which is below today's regulation (100 mg Cd kg<sup>-1</sup> P), would also lead to a slow accumulation if maximum allowed application of sludge is used. With ML level 91.6 and 137.4 mg Cd kg<sup>-1</sup> P, it is predicted a 4 and 11% increase, respectively, in 100-year perspective. This also indicates that at Ås, use of mineral fertilisers with Cd concentration up to today's Norwegian regulation (100 mg Cd kg<sup>-1</sup> P) together with maximum allowed sewage sludge with an average Cd concentration (0.6 mg Cd kg<sup>-1</sup> DW, might cause a slow increase and not decrease over time.

The effects of different Cd contents in mineral P fertilisers is initially insignificant, but increases with time. The biggest increase is found in Melhus, where the PEC and RCR after 100 years using fertilisers with the highest content of Cd in mineral P are 68% higher than

using fertilisers with the present content of Cd in mineral P for 100 years. However, the RCR is still well below 1, indicating no risk of effects on terrestrial organisms.

The same trend as predicted in this risk assessment is also recently reported in an EU risk assessment. A combination of lower Cd input to arable soil and previous overestimation of Cd sorption to soil, probably explain the trend with declining Cd concentration over time in soil, which differs from former risk assessments. This possible underestimation of removal from soil, call for more monitoring of Cd in water monitoring programs. It is also important to be aware of that some areas might accumulate Cd in soil over time. Climate change, e.g. increased annual precipitation, and change in pH and soil organic matter, will influence the fate of Cd in the environment, and, thus, the transfer pathway of Cd in the environment.

### 3. What level of cadmium in agricultural soils would give the risk of negative effects on the affected organisms specified in Table A?

Predicted No Effect Concentrations (PNEC) for different categories of aquatic and terrestrial organisms have been adopted from the European Risk Assessment Report on Cd (EC 2007) and are as follows:

<u>Terrestrial organisms</u>, <u>direct effect</u>: For microbial communities and invertebrates inhabiting soil, negative effects may occur at Cd concentrations in soil above the PNEC<sub>soil</sub> =  $2.3 \text{ mg Cd kg}^{-1}$  DW.

<u>Agricultural plants:</u> Higher plants may be negatively affected at Cd concentrations in soils above the  $PNEC_{soil\ plants} = 2.8 \text{ mg Cd kg}^{-1} DW$ .

<u>Terrestrial organisms secondary poisoning:</u> Mammals may be affected by secondary poisoning of Cd from ingesting plants

or prey in areas with soil Cd concentrations above PNEC<sub>soil, secondary poisoning</sub>= 0.9 mg Cd kg<sup>-1</sup> DW.

<u>Aquatic organisms</u>: Aquatic organisms may be affected at concentrations of dissolved Cd at concentrations in water above the PNEC<sub>water</sub> =  $0.08 \mu g L^{-1}$ .

<u>Sediment dwelling (benthic) organisms</u>: Benthic organisms dwelling on or in freshwater sediments may be affected at Cd concentrations in sediment above the  $PNEC_{sediment} = 2.3 \text{ mg}$  Cd  $kg^{-1}$  DW.

<u>Farm animals:</u> The estimated Cd exposure of farm animals consuming feed and/or grass from fields where mineral fertilisers have been used will mainly decrease, and the exposure is considered far lower than those at which toxicological data indicate adverse effects will occur. However, Cd may accumulate in the liver and kidneys but probably not above the

maximum limits in these tissues for human consumption. At all regions the estimated Cd exposure is below, the upper limit set by WHO of 1 mg Cd  $kg^{-1}$  dried feed, which protects the animal health, but may, over time, probably result in Cd in liver and kidney above regulated limits.

- 4. How will application of mineral phosphorus fertilisers with a cadmium content of 137.4, 91.6, or 45.8 mg Cd kg<sup>-1</sup> phosphorus under different crop rotations:
  - a. affect the levels of cadmium in agricultural soil in Norway from a 1, 10, and 100-year perspective.
  - b. affect the risk of negative effects on the target organisms specified in Table A?

### a) The effect on Cd levels in agricultural soils in Norway from a 1, 10, and 100year perspective.

As described under question 2, declines in concentrations of Cd in soil over time at the three municipalities Time, Melhus, and Stange are predicted. This means that the current background concentration of Cd in soil is higher than the predicted soil concentrations in 1, 10, and 100 years' time. The same trend of decreasing soil concentrations has been reported by Six and Smolders (2014) in their evaluation of the effect of phosphorus fertilisers on Cd accumulation in European agricultural soils.

At Ås, the situation was different. Using the chosen soil quality parameters, annual precipitation and infiltration rates, and a realistic worst-case sewage sludge application, ML values of 91.6 and 137.4 mg Cd kg<sup>-1</sup> P, showed an estimated accumulation of Cd in soil from the long-term perspective. The contribution of Cd via sewage sludge was the key factor in this case. Under the same conditions, but with use of an average amount of sewage sludge application for the municipality (based on data from SSB), the total input of Cd to soil decreased, altering the Cd mass balance in soil, and resulting in a net Cd removal and a predicted reduction of concentrations of Cd in soil in the long term.

### b) The effect on risk of negative effects on target organisms.

The risk is expressed quantitatively by Risk Characterisation Ratio (RCR), defined as the ratio between PEC and PNEC.

#### **Terrestrial animals:**

• The RCRs are below 1 for all scenarios indicating no risk from direct exposure to soil for terrestrial organisms. The highest RCRs (0.74) are found in Stange where the present background Cd concentration is 26 % below the PNEC<sub>soil</sub>.

- The RCRs indicate a risk of secondary poisoning of terrestrial mammals and birds at the present level of CD in soil at Stange. The concentrations are predicted to decline with time, but the RCRs are still above 1 after 100 years, independent of the level of Cd in fertilisers. For the Ås, Time, and Melhus scenarios, all RCRs are well below 1 indicating no risk of secondary poisoning.
- The effect of different Cd contents in fertilisers is the same as described for agricultural plants above.

#### Agricultural plants:

- The RCR are below 1 for all scenarios, indicating no risk from direct exposure to soil for terrestrial plants, even at the highest Cd content fertilisers (137.4 mg Cd kg<sup>-1</sup> P).
- In most scenarios, the PECs, and consequently also the RCRs, will decline over 100 years. The effects of different Cd concentrations in mineral P fertilisers are initially insignificant, but increase with time. The biggest increase is found in Melhus, where the PEC and RCR after 100 years of use of the highest content of Cd in mineral P are 68% higher than after 100 years of use with fertilisers with the present content of Cd in mineral P. The RCR is, however, still well below 1, indicating no risk of effects on agricultural plants.

### Aquatic organisms:

• The estimated concentrations of Cd in surface water indicate that current background concentrations of Cd in agricultural soils may constitute a risk of effects on aquatic organisms in recipients of drainage water from arable land. This is particularly the case in the alum shale area (Stange), where RCR is 3, based on the present background concentration. In Time, the RCR is 0.97 but decreases to 0.21 after 100 years with use of fertilisers with the highest content of Cd. In Ås, where Cd concentrations increase slightly with use of fertilisers with the two highest Cd-levels, the RCR increases from 0.69 to 0.75 with 137.4 mg Cd kg<sup>-1</sup> P.

#### <u>Sediment-dwelling (benthic) organisms</u>

• The RCR at Stange is 1, indicating a risk to sediment-dwelling organisms with the background concentration of Cd in soil, and after one year with fertiliser application. But as the concentrations of Cd decrease with time, the RCRs are below 1 after 10 and 100 years, indicating no further risk of effects.

Farm animals for food production, grazing where mineral fertilisers have been used:

The model for Cd concentrations in feed and pasture plants, even using fertiliser with the highest level of Cd for 100 years (worst case), found that Cd levels will mainly decline. Thus, Cd exposure to farm animals is not expected to increase from the present exposure level.

In all selected geographical regions, including the alum shale area, current Cd levels in feed and pasture plants, and thus farm animal diets, are below a critical level of concern for consumers of animal products and far below the level of animal health concern.

By using mineral fertiliser, Cd concentrations in animal diets will mostly decrease and thus reduce the risk of reaching Cd concentrations of concern in animal products, as well as the less likely risk for adverse health effects in animals. However, prevention of liver and kidneys of older animals from entering the human food chain remains a measure to decrease the consumer exposure to Cd.

Cd levels in animal drinking water (from the tap) is usually far below that in their feed and pasture plants, and is not expected to contribute significantly to the total exposure.

5. How will mineral phosphorus fertilisers with a cadmium content of 137.4, 91.6, or 45.8 mg Cd kg<sup>-1</sup> phosphorus affect the dietary exposure to cadmium for Norwegians in general and for subgroups of the population in a 1, 10, and 100-year perspective?

None of the Cd concentrations used in the modelling will have any impact on the dietary exposure to Cd after 1 year. From a longer perspective, Cd concentrations in crop plants will based on the predictions decrease in three of the municipalities, and hence also the dietary exposure to Cd will decrease, albeit with only very minor differences in the decrease between the different Cd concentrations in fertilisers. The median effects will depend on various factors, including the proportion of plants cultivated in soil represented by the different scenarios, and have not been estimated for other crops, but would probably be in the range between the lowest and highest decrease. At Ås, a slight increase in Cd concentrations in crops was predicted from a 100-year perspective.

The current mean dietary exposure of Cd is estimated to exceed the TWI for the youngest age groups and for most age groups for the high consumers (see Table 7.3.3.2-1), and a reduction in Cd exposure is therefore desirable as this will reduce the health risk in the population. A reduction in Cd concentrations in the main dietary sources such as cereals, potatoes, and root vegetables is therefore likely to decrease exposure, and hence reduce the risk for the population.

VKM has evaluated risk for children eating soil, taking into account that this exposure route usually only occurs in a limited age range. Given the low estimated exposure compared with intake from the diet, this exposure route is considered to be of low risk.

### 12 Data gaps

- At present, there are no regional or national harmonized datasets available that provide the natural concentration levels of Cd - or other potentially toxic elements (PTE)- in Norwegian agricultural soil. The Cd concentrations in agricultural soil used in this risk assessment have been obtained from several independent projects, where they have been collected without any harmonized procedures (agricultural soil, soil depth, sampling preparation practice, analytical techniques), and many of the data back to 1996. A monitoring programme/campaign for Cd and other PTEs in Norway would help to decrease uncertainty for further risk assessments.
- Bioavailability, mobility, and aging of geogenic and anthropogenic Cd (as well as other PTE with geogenic presence) in Norwegian soil types, particularly alum shale soil (e.g., Stange) should be studied in order to obtain better predictions of environmental fate and transfer to crops and further exposure of farm animals and humans, and also for consideration of effects on terrestrial predators (secondary poisoning).
- Monitoring campaigns for Cd in mineral fertiliser for Cd concentration should be performed in order to verify the actual Cd concentration.
- Cd analyses should be included in monitoring programme for groundwater (including private drinking water wells) and surface water, including lakes, as well in programmes or other larger projects funded by the authority.
- As it has recently been recognized that Kd values have been used for predicting fate of Cd
  in soil are too high, followed by an underestimate of the leaching processes, there is reason
  to investigate the impact that this has on aquatic and sediment-dwelling organisms.

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# Appendix I

## **Background Cd soil concentrations**

In previous Norwegian risk assessments regarding Cd, the number of background soil samples have been few in number (as low as n=4). Thus, at the start of this risk assessment, VKM undertook an active search for background data from a range of sources. The data on soil concentrations of Cd used in this report have been obtained from Reimann et al., 2014; Reimann et al., 2003; Fylkesmannen in Vestfold, 2000; Esser, 1996; NIBIO (Joner, unpublished data); Dragievic, 2015, 2016.

**Table A I-1a:** Cadmium concentrations in agricultural soil from different municipalities in Southwestern Norway. References (Ref.): 1: Reimann et al., 2014 (arable land); 2: Reimann et al., 2014 (grazing land); 4: Esser, 1996 (arable land); 6: Reimann et al., 2003 (arable land).

Cd				Cd	
Municipality	mg/kg	Ref.	Municipality	mg/kg	Ref.
Audnedal	0.61	2	Sola	0.03	4
Audnedal	0.23	1	Sola	0.05	4
Audnedal	0.19	6	Songdalen	0.36	2
Birkenes	0.22	4	Songdalen	0.30	1
Bjerkreim	0.33	2	Songdalen	0.27	6
Farsund	0.36	2	Suldal	0.13	6
Farsund	0.20	1	Suldal	0.03	4
Flekkefjord	0.26	6	Time	0.25	2
Flekkefjord	0.18	6	Time	0.33	1
Haugesund	0.32	2	Time	0.21	6
Haugesund	0.52	1	Time	0.16	4
Hjelmeland	0.40	2	Time	0.13	4
Hjelmeland	0.22	1	Vindafjord	0.12	2
Hjelmeland	0.15	6	Vindafjord	0.11	1
Iveland	0.11	4	Vindafjord	0.36	1
Karmøy	0.15	4	Vindafjord	0.19	6
Kvinesdal	0.17	4	Vindafjord	0.14	4
Kvinesdal	0.33	4			
Lyngdal	0.18	2			
Lyngdal	0.16	1			
Lyngdal	0.23	4			
Lyngdal	0.12	4			
Sirdal	0.32	2			
Sirdal	0.18	2			
Sirdal	0.17	1			
Sirdal	0.13	1			

**Table A I-1b:** Cadmium concentrations in agricultural soil from different municipalities in Trøndelag county. References (Ref.): 1: Reimann et al., 2014 (arable land); 2: Reimann et al., 2014 (grazing land); 4: Esser, 1996 (arable land); 6: Reimann et al., 2003 (arable land).

Cd				Cd	<u></u>
Municipality	mg/kg	Ref.	Municipality	mg/kg	Ref.
Grong	0.13	2	Rissa	0.16	4
Grong	0.09	1	Rissa	0.04	4
Grong	0.12	6	Rissa	0.10	4
Hemne	0.06	2	Røros	0.03	2
Hemne	0.10	1	Røros	0.03	1
Hemne	0.04	6	Røros	0.19	6
Hitra	0.10	2	Røyrvik	0.20	2
Hitra	0.15	1	Røyrvik	0.11	1
Leksvik	0.02	4	Røyrvik	0.10	6
Leksvik	0.10	4	Selbu	0.27	2
Levanger	0.11	2	Selbu	0.14	1
Levanger	0.16	1	Selbu	0.10	4
Levanger	0.09	6	Selbu	0.08	4
Lierne	0.12	2	Snåsa	0.13	4
Lierne	0.08	2	Steinkjer	0.17	2
Lierne	0.20	1	Steinkjer	0.11	1
Lierne	0.20	1	Steinkjer	0.12	6
Lierne	0.19	6	Steinkjær	0.05	4
Melhus	0.08	6	Steinkjær	0.08	4
Melhus	0.17	4	Steinkjær	0.10	4
Melhus	0.14	4	Steinkjær	0.15	4
Mosvik	0.06	4	Stjørdal	0.10	4
Namsos	0.06	2	Stjørdal	0.11	4
Namsskogan	0.14	2	Stjørdal	0.05	4
Namsskogan	0.10	1	Stjørdal	0.07	4
Namsskogan	0.07	6	Trondheim	0.11	4
Oppdal	0.12	6	Trondheim	0.06	4
Orkdal	0.04	1	Trondheim	0.08	4
Orkdal	0.08	6	Tydal	0.11	6
Overhalla	0.04	1	Verdal	0.10	2
Overhalla	0.14	6	Verdal	0.13	1
Rennebu	0.16	2	Verdal	0.11	6
Rennebu	0.05	2	Verdal	0.13	4
Rennebu	0.11	1	Verdal	0.10	4
Rennebu	0.09	6	Verdal	0.08	4
Rissa	0.16	2	Verdal	0.08	4
Rissa	0.11	1	Åfjord	0.08	2
Rissa	0.18	6	Åfjord	0.09	1
Rissa	0.10	4	Åfjord	0.09	6

**Table A I-1c:** Cadmium concentrations in agricultural soil from different municipalities in Hedmark county. References (Ref.): 1: Reimann et al., 2014 (arable land); 2: Reimann et al., 2014 (grazing land); 5: NIBIO (Joner, unpublished data); 6: Reimann et al., 2003 (arable land); 7: Joner, NIBIO, personal communication and unpublished data from ongoing project, 2018.

personal commi	Cd	ia aripabile		Cd	n Ojece,	2010.	Cd	
Municipality	mg/kg	Ref.	Municipality	mg/kg	Ref.	Municipality	mg/kg	Ref.
Elverum	0.04	2	Stange	2.30	5	Tolga	0.08	1
Elverum	0.05	1	Stange	2.30	5	Tolga-Os	0.22	4
Elverum	0.15	6	Stange	2.70	5	Tolga-Os	0.21	4
Elverum	0.12	4	Stange	1.30	5	Trysil	0.14	2
Elverum	0.34	4	Stange	1.10	5	Trysil	0.08	1
Engerdal	0.14	2	Stange	1.70	5	Trysil	0.16	6
Engerdal	0.04	2	Stange	0.90	5	Tynset	0.05	2
Engerdal	0.08	1	Stange	1.80	5	Tynset	0.09	1
Engerdal	0.02	1	Stange	0.77	5	Tynset	0.21	6
Engerdal	0.06	6	Stange	0.68	5	Tynset	0.14	6
Engerdal	0.08	6	Stange	0.44	5	Åmot	0.08	2
Folldal	0.19	2	Stange	0.39	5	Åmot	0.05	1
Folldal	0.19	2	Stange	0.84	5	Åmot	0.06	6
Folldal	0.16	1	Stange	0.85	5	Åsnes	0.20	2
Folldal	0.13	1	Stange	1.30	4	Åsnes	0.15	1
Folldal	0.10	6	Stange	1.80	4	Åsnes	0.09	6
Hamar	0.34	5	Stange	2.57	7			
Hamar	0.57	5	Stange	3.26	7			
Hamar	0.55	5	Stange	1.96	7			
Hamar	0.71	5	Stange	1.35	7			
Hamar	0.54	5	Stange	2.37	7			
Hamar	0.62	5	Stange	2.19	7			
Hamar	1.10	5	Stange	0.42	7			
Hamar	0.40	5	Stange	0.82	7			
Hamar	0.22	5	Stange	1.58	7			
Hamar	0.25	5	Stange	2.25	7			
Kongsvinger	0.08	1	Stange	1.66	7			
Kongsvinger	0.06	6	Stange	2.05	7			
Ringsaker	0.24	1	Stange	2.27	7			
Ringsaker	0.32	6	Stange	2.42	7			
Ringsaker	0.18	4	Stange	3.28	7			
Ringsaker	0.17	4	Stange	2.97	7			
Stange	0.73	2	Stange	3.84	7			
Stange	0.81	1	Stange	2.71	7			
Stange	1.40	5	Stor-Elvdal	0.07	2			
Stange	1.10	5	Stor-Elvdal	0.17	1			
Stange	2.80	5	Stor-Elvdal	0.17	6			
Stange	0.76	5	Stor-Elvdal	0.13	6			
Stange	0.90	5	Tolga	0.08	2			

**Table A I-1d:** Cadmium concentrations in agricultural soil from different municipalities in Southeastern Norway (Østlandet). References (Ref.): 1: Reimann et al., 2014 (arable land); 2: Reimann et al., 2014 (grazing land); 3: Fylkesmannen in Vestfold, 2000 (arable land); 4: Esser, 1996 (arable land); 5: NIBIO (Joner, unpublished data); 6: Reimann et al., 2003 (arable land); 7: Joner, NIBIO, personal communication and unpublished data from ongoing project, 2018; 8: Dragievic, 2015, 9: Dragievic, 2016.

9: Dragievic, 2016.	Cd			Cd			Cd	
Municipality	mg/kg	Ref.	Municipality	mg/kg	Ref.	Municipality	mg/kg	Ref.
Andebu	0.20	2	Ramnes	0.21	3	Sandefjord	0.60	3
Andebu	0.24	1	Ringerike	0.17	4	Sandefjord	0.50	3
Aurskog-Høland	0.21	2	Ringerike	0.12	4	Sandefjord	0.50	3
Aurskog-Høland	0.09	1	Sandefjord	0.11	3	Sandefjord	0.60	3
Borre	0.13	3	Sandefjord	0.20	3	Sandefjord	0.60	3
Borre	0.11	3	Sandefjord	0.18	3	Sandefjord	0.60	3
Borre	0.25	3	Sandefjord	0.20	3	Sandefjord	0.12	3
Eidsvoll	0.21	4	Sandefjord	0.22	3	Sandefjord	0.12	3
Eidsvoll	0.18	4	Sandefjord	0.14	3	Sandefjord	0.30	3
Enebakk	0.28	2	Sandefjord	0.30	3	Sandefjord	0.44	3
Enebakk	0.28	1	Sandefjord	0.17	3	Sandefjord	0.28	3
Fredrikstad	0.33	2	Sandefjord	0.26	3	Sandefjord	0.13	3
Fredrikstad	0.15	1	Sandefjord	0.24	3	Ski	0.22	6
Gran	0.29	4	Sandefjord	0.13	3	Skien	0.15	4
Gran	0.30	4	Sandefjord	0.14	3	Skien	0.13	4
Grue	0.11	4	Sandefjord	0.14	3	Skien	0.38	4
Grue	0.10	4	Sandefjord	0.14	3	Trøgstad	0.21	4
Hof	0.19	2	Sandefjord	0.12	3	Trøgstad	0.18	4
Hof	0.26	1	Sandefjord	0.07	3	Tønsberg	0.16	3
Hof	0.11	6	Sandefjord	0.02	3	Våle	0.02	3
Holmestrand	0.21	3	Sandefjord	0.45	3	Våle	0.08	3
Holmestrand	0.08	3	Sandefjord	0.21	3	Våle	0.33	4
Hurdal	0.19	2	Sandefjord	0.27	3	Våle	0.32	4
Hurdal	0.20	1	Sandefjord	0.22	3	Våler	0.36	2
Hurdal	0.26	6	Sandefjord	0.40	3	Våler	0.16	2
Larvik	1.00	3	Sandefjord	0.26	3	Østre Toten	1.60	4
Lier	0.22	4	Sandefjord	0.23	3	Østre Toten	0.81	4
Lier	0.19	4	Sandefjord	0.30	3	Ås	0.14	8
Løten	0.25	4	Sandefjord	0.31	3	Ås	0.16	9
Løten	0.17	4	Sandefjord	0.19	3	Ås	0.13	5
Nittedal	0.16	2	Sandefjord	0.40	3	Ås	0.19	5
Nittedal	0.28	1	Sandefjord	0.10	3	Ås	0.19	5
Nittedal	0.24	6	Sandefjord	0.70	3			
Rakkestad	0.16	2	Sandefjord	0.50	3			
Rakkestad	0.22	1	Sandefjord	0.30	3			
Ramnes	0.34	3	Sandefjord	0.30	3			
Ramnes	0.19	3	Sandefjord	0.42	3			
Ramnes	0.18	3	Sandefjord	0.44	3			

The number of samples in the four regions ranges from 43 in southwestern Norway to 108 from southeastern Norway. The number of analyses from the participating municipalities also varies, from 1 value up to as many as 47 (Sandefjord, southeastern Norway). The distribution of data from the different municipalities in the four regions is summarised in Table A I-2.

**Table A I-2:** Number of samples from each municipality from the four regions. N is the total number

of samples from the region.

Southwestern	Norway	Hedmar	k	Trøndela	ıg	Southeastern N	orway
Municipality	N=43	Municipality	N=94	Municipality	N=78	Municipality	N=108
Audnedal	3	Elverum	5	Grong	3	Andebu	2
Birkenes	1	Engerdal	6	Hemne	3	Aurskog-Høland	2
Bjerkreim	1	Folldal	5	Hitra	2	Borre	3
Farsund	2	Hamar	10	Leksvik	2	Eidsvoll	2
Flekkefjord	2	Kongsvinger	2	Levanger	3	Enebakk	2
Haugesund	2	Ringsaker	4	Lierne	5	Fredrikstad	2
Hjelmeland	3	Stange	41	Melhus	3	Gran	2
Iveland	1	Stor-Elvdal	4	Mosvik	1	Grue	2
Karmøy	1	Tolga	2	Namsos	1	Hof	3
Kvinesdal	2	Tolga-Os	2	Namsskogan	3	Holmestrand	2
Lyngdal	4	Trysil	3	Oppdal	1	Hurdal	3
Sirdal	4	Tynset	4	Orkdal	2	Larvik	1
Sola	2	Åmot	3	Overhalla	2	Lier	2
Songdalen	3	Åsnes	3	Rennebu	4	Løten	2
Suldal	2			Rissa	7	Nittedal	3
Time	5			Røros	3	Rakkestad	2
Vindafjord	5			Røyrvik	3	Ramnes	4
				Selbu	4	Ringerike	2
				Snåsa	1	Sandefjord	47
				Steinkjer	3	Ski	1
				Steinkjær	4	Skien	3
				Stjørdal	4	Trøgstad	2
				Trondheim	3	Tønsberg	1
				Tydal	1	Våle	4
				Verdal	7	Våler	2
				Åfjord	3	Østre Toten	2
						Ås	5

The minimum, mean, median, and maximum Cd concentrations from each region, based on all data provided in Tables A Ia-Id, are summarised in Table A I-3. It should be noted that, the data from the regions Hedmark and southeastern Norway are dominated by the large number of analyses from the municipalities of Stange and Sandefjord, respectively. In risk

assessment analyses of the entire region, only minimum, mean, median, and maximum Cd concentration values are used from these two municipalities (Table A I-4). A statistical summary of Cd concentrations for each region after 'smoothing out' the data from Stange and Sandefjord is given in Table A I-5.

Boxplots of all data and with 'smoothed' data from Stange and Sandefjord are provided in Fig. A I-1.

**Table A I-3** Statistical summary of all data given in Table A I-1a-1d.

Region	MIN	MEAN	MEDIAN	MAX
	mg/kg	mg/kg	mg/kg	mg/kg
Hedmark	0.015	0.86	0.395	3.8
Southwestern Norway	0.025	0.22	0.19	0.61
Trøndelag	0.022	0.11	0.10	0.27
Southeastern Norway	0.020	0.26	0.21	1.6

<sup>\*</sup>Number of samples in each region is given in Table A I-1a-1d.

**Table A I-4:** Statistical summary of data from focus area Stange (Hedmark) and

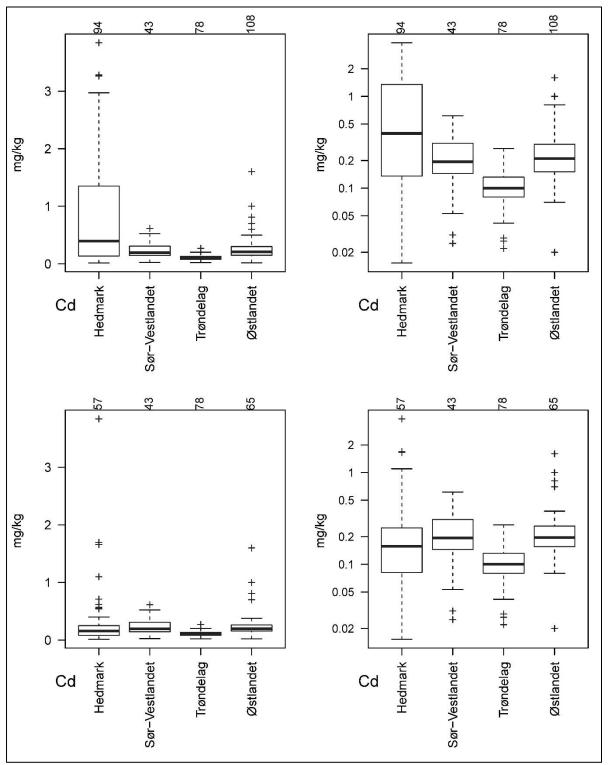
Sandefjord (Southeastern Norway)

ouriacijora (ooaa	ioastoiii itoiitaj			
	MIN	MEAN	MEDIAN	MAX
Municipality	mg/kg	mg/kg	mg/kg	mg/kg
Sandefjord	0.02	0.29	0.26	0.7
Stange	0.39	1.7	1.7	3.8

**Table A I-5:** Statistical summary for each region after 'smoothing out' the data from Stange and Sandefjord by using only minimum, mean, median, and maximum

values from these two municipalities.

values from these two mans	MIN	MEAN	MEDIAN	MAX
Region	mg/kg	mg/kg	mg/kg	mg/kg
Hedmark	0.015	0.32	0.16	3.8
Southwestern Norway	0.025	0.22	0.19	0.61
Trøndelag	0.022	0.11	0.10	0.27
Southeastern Norway	0.020	0.25	0.20	1.6



**Fig. A I-1.** Boxplots of all data from four Norwegian regions. Please note that the right-hand figure has a log scale while the left-hand figure has not. The number of samples in each region is given on the top axis.

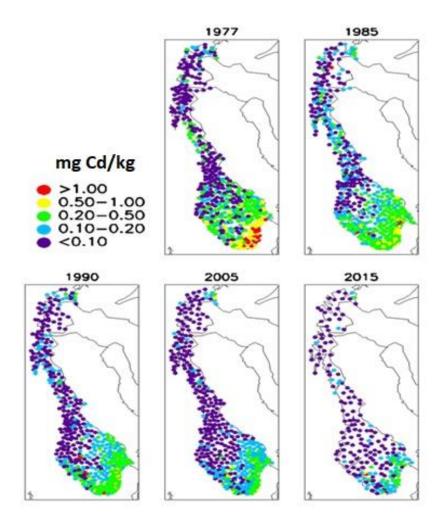
Top left and top right are boxplots of all data from four Norwegian regions, compiled from the dataset in Table A I-1a-1d.

Bottom left, and bottom right are boxplots of all data, but data from Stange and Sandefjord have been smoothed. Both right-hand plots have a log scale, but the left-hand figures do not. The number of samples in each region is given on the top axis of all plots.

## Appendix II

## **Atmospheric Cd deposition data from Norway**

During the 1970s it became evident that the southernmost part of Norway was considerably affected by air pollutants originating from other parts of Europe. Substantial efforts have been made to study the extent of this pollution: source areas, geographical distribution within Norway, and possible effects. Whereas the emphasis during the first period was on acidic deposition and its effects on freshwater ecosystems, the establishment of air monitoring programmes and development of gradually improved analytical techniques revealed that some PTEs were also among the pollutants transported into Norway and supplied to terrestrial ecosystems by atmospheric deposition.



**Fig. A II-1.** Decrease in atmospheric deposition of Cd in Norway from 1977 to 2015, illustrated by Cd concentrations in moss samples, units in mg Cd kg<sup>-1</sup> moss (Steinnes et al., 2016).

Elements supplied by this route are mostly those that may be released in volatile form from high-temperature sources, such as burning fossil fuel, metallurgical processes, and automobile engines. Among these elements, most attention has been directed towards lead and mercury, but several other elements, such as zinc, arsenic, cadmium, and antimony, are also supplied from this general source. Continental-scale monitoring, based on sampling of terrestrial moss according to an extensive network and subsequent chemical analyses, has revealed the geographic distribution and temporal trend of this metal pollution in Norway (Steinnes et al., 2016) and, more recently, in large parts of Europe (Harmens et al., 2010).

Monitoring of bulk deposition of pollutants is carried out regularly by sampling and analysis of precipitation samples, but only at a few sites in different parts of Norway (Table A II-1). Since around 1990, this monitoring has included metal pollutants such as Cd, Pb, and Zn. Introduction of moss sampling and sufficiently sensitive analytical techniques have facilitated much more detailed knowledge about the temporal and spatial trends in atmospheric deposition of these elements. The moss survey for deposition of PTEs was introduced in 1977, covering around 460 sites all over Norway, and, since then, has been carried out every 5 years.

The temporal and spatial trends of Cd deposition in Norway are illustrated in Fig. A II-1 (Steinnes et al., 2016). The atmospheric deposition of Cd and other metals shows a distinct geographical trend, with the highest levels in the southernmost parts of the country. However, it has substantially reduced over time and is currently not significant. As evident from Fig. A II-1, the Cd deposition level during the mid-1970s was about 10 times higher in the southernmost parts of Norway than in areas least exposed to transboundary pollution from Europe (Dovre region and Indre Troms/Vest-Finnmark). The drastic reduction in atmospheric deposition of Cd observed in moss during the last 20 to 30 years is in consensus with monitoring data from the rural air- and precipitation chemistry monitoring network in Norway (Table A II-1) and Denmark (Aase et al. 2012; Bohlin-Nizzetto et al., 2017; Ellermann et al., 2015).

**Table A II-1.** Atmospheric Cd contributions from monitoring data from the rural air- and precipitation chemistry monitoring network in Norway (data from 2006, 2012 and 2017 shown).

SHOWITH			
Total wet deposition of Cd	2006 <sup>1</sup>	2012¹	2017 <sup>2</sup>
Birkenes	0.496	0.338	0.360
Hurdal	0.478	0.198	0.120
Kårvatn	0.078	0.068	0.050
Svanvik	0.602	0.17	0.360
Average	0.414	0.194	0.223

<sup>&</sup>lt;sup>1</sup> Aase et al. 2012

## Appendix III

#### **Selection of Kd**

#### A III-1. Previously used Kd-regressions

Kd is a critical parameter for long-term simulations in environmental and health risk assessment of pollutants. Selection of Kd for this risk assessment, as well as other risk assessments performed for Cd, is therefore described here.

The report "Risk assessment of Cd in mineral fertilisers in Norway using model calculations" from Amundsen et al. (2000) uses the Kd regression from McBride et al. (1997). The same authors (i.e., Sauvé et al., 2000) have since published a new regression. In the risk assessment of Smolders (2013) "Revisiting and updating the effect of phosphorous fertilisers on cadmium accumulation", another and new regression is used, merged from four previous regressions.

In Amundsen et al. (2000), the Kd-regression of McBride et al. (1997) was used:

$$log C_water = 3.62 - 0.5 pH + 0.96 log C_soil - 0.45 log OM$$
 (eq. A III-1)

where

C\_water = Cd concentration in soil pore water (mg/L)
C\_soil = Cd concentration in dry weight soil (mg/kg DW)
OM = organic matter (g/kg)

Kd is then the ratio of C\_soil to C\_water (L/kg).

The Kd-regression derived by Smolders (2013) is:

$$log Kd = -0.94 + 0.51 pH + 0.79 log (\%OC)$$
 (eq. A III-2)

where %OC is organic carbon (% weight based)

In Sauve et al. (2000), two regressions for the Kd of Cd were listed. Only the one with pH and OM as input parameters is compared here, because it has the higher R<sup>2</sup>:

$$log Kd = 0.48 pH + 0.82 log SOM - 0.65$$
 (eq. A III-3)

In order to compare the Kd equations from Christensen (1989), Sauve (2000), Römkens and Salomon (1998), McBride and Sauve (1997), and Smolders 2013, these were tested for pH 4, 5.5, and 7, and SOM 2, 10, and 25.

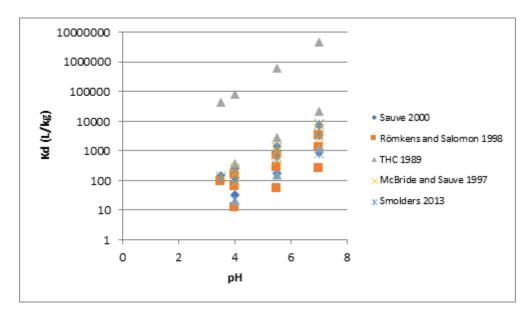
The following Fig. A III-1 shows a comparison for pH = 4, 5.5, and 7, and for OM = 2, 10, and 25% (plus one soil pH 3.5 and OM 25%) for all regressions listed above. It can immediately be seen that the method of Christensen (1989) is slightly higher than the others (for OM = 25%), and Norwegian soils are out of the range of applicability of this regression.

In Fig. A III-2, the Kd-regression of Christensen (1989) is therefore omitted. It is then clearer that the Kd of McBride et al. (1997) is, in all cases and at all OM, the highest Kd of all estimates.

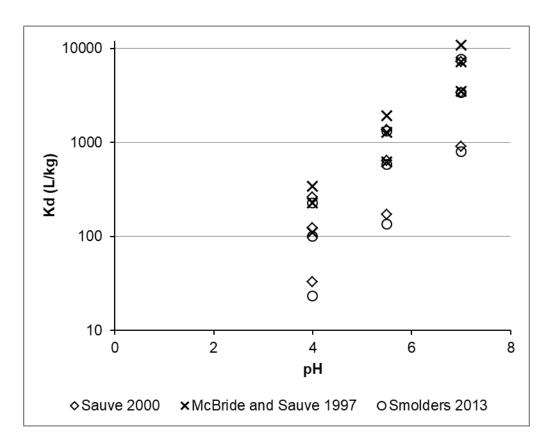
The regression of Römkens and Salomon (1998) gives the lowest Kd, but the Sauvé (2000) regression and the regression derived from Smolders (2013) are in quite close agreement.

#### A III-2. Comparison of results

In order to compare the Kd-equations, these were tested for pH of 4, 5.5, and 7, and for soil organic matter OM of 2, 10, and 25 g C/g (conversion 1.724 g OM is 1 g OC). Fig. A III-1 shows the comparison of the three Kd-equations. It can be seen that the Kd calculated using the equation of Sauvé et al. (2000) and that derived independently by Smolders (2013) from four other data sets give very similar results, whereas the equation of McBride et al. (1997) is higher than the two others throughout, and, at the lowest pH and OM factor, 4.7 times higher.



**Fig. AIII-1:** Comparison of Kd-equations for pH of 4, 5.5, and 7 and soil organic matter of 2, 10, and 25 g C/g (conversion 1.724 g OM is 1 g OC).



**Fig. A III-2:** Comparison of Kd-equations for pH of 4, 5.5, and 7 and soil organic matter of 2, 10 and 25 g C/g (conversion 1.724 g OM is 1 g OC).

#### A III-3. Conclusions and consequences

With simulations over 100 years, a high Kd would predict higher accumulation in soils. On the other hand, the immediate (now-time) risk for leaching and plant uptake is lower. The regression used to date in risk assessment of Cd in Norwegian soils is from McBride et al. (1997) and gives relatively high Kd-values. **High Kd-values in the model algorithm will lead to a predicted high accumulation of Cd in soils over long-term simulations and thus, in the long run, will predict a higher potential risk.** Two good alternatives to this regression are the scientifically well-established regression of Sauvé et al. (2000) and the more recently established regression by Smolders et. al (2013).

Due to the very close agreement between the regression of Smolders and that of Sauvé, both are considered reliable. An additional argument that favours the Smolders regression is that it allows direct comparison of this study with the recent risk assessment of Cd in phosphate fertiliser (Smolders, 2013).

## Appendix IV

### Evaluation of methods for transfer to crops and animal feed

Plant uptake of Cd depends on soil properties, availability of Cd in soil, and on properties of the plant itself (McLaughlin et al., 2011). Cd is a non-essential metal and therefore most likely taken up passively (i.e., with the soil water). A couple of methods for predicting uptake of Cd into crops have been derived. Most of these methods are regression equations or TF (ERM, 2000; Smolders, 2013; Legind and Trapp, 2010, Novotná et al., 2015). A mechanistic model has also been developed (Legind et al., 2012). More complex regressions use multiple predictor variables (typically pH, organic C, clay content) (Novotná et al., 2015). In order to find the most accurate and least data-intensive prediction method, we compared three methods for conditions in Norway with measured Cd data in Norwegian crops.

#### i) Transfer factors (TF) (Smolders, 2013)

TF values of Smolders are defined as TF = C(crop) / C(soil),

where C(crop) is in mg/kg FW, and C(soil) is in mg/kg (usually DW, but not given). TF values were derived from average concentrations in several European countries, listed in ECB (2007), Table 3.182 page 285.

TF (wheat) = 0.14 mg/kg wheat FW / mg kg soil

TF (potato) = 0.06 mg/kg potato FW / mg kg soil

#### ii) Regression to soil pH and soil Cd concentration (ERM 2000)

The approach used in Amundsen et al. (2000) and recommended by ERM (2000) is based on regressions in Erickson et al. (1996) using constant values for soil organic matter and zinc concentrations in soil.

BCF regressions for Cd uptake into winter wheat, potatoes and carrots

```
C(winter wheat) = 81 - 10.3 pH + 100 C(soil) (mean zinc = 42.1 mg/kg)
```

C(potatoes) = 177-24.1 pH + 39 C(soil) (mean OM = 17.3%)

C(carrots) = 1289 - 165 pH

where C(plant) is the Cd concentration in plants (presumably DW, but not given in the report) and C(soil) is the Cd concentration in soil (presumably DW).

#### iii) Mechanistic model (Legind et al. 2012)

Non-essential elements like Cd are taken up passively into plants (McLaughlin et al., 2009). Then, mass of Cd in plant = Cd uptake by translocation plus deposition from air

= Q x Csoil / Kd + aerial deposition x interception

Transpiration Q (L) is precipitation minus excess precipitation and was taken from Amundsen et al. (2000), Table 7 (alternatively it may be set to 400 L/m²/growth season)

Aerial deposition in 2018 was (on average)  $0.08 \text{ g/ha/a} = 8 \mu\text{g/m}^2/\text{a}$ 

(for comparison with measured data, the average 1995 to 1998 was used, see Amundsen et al. Table 12; overall, uptake of Cd into plants due to deposition from air is negligible for Norwegian conditions)

Interception by plants is from 0% (winter) to 75% (before harvest) and was set to an average of 25%.

Kd is estimated with the regression of Smolders (2013): log Kd = 0.51 pH + 0.79 log %OC - 0.94

Concentration in plant is mass of Cd / mass of plants

Mass of plant was set to 10 kg FW at harvest (total plant, harvest product plus remainder plus roots), all data per m<sup>2</sup>. The method allows use of plant-specific data, but those were not available. This mechanistic model has been used previously and shown good results (Legind et al., 2012).

#### **Method comparison**

The methods were compared for six Norwegian regions. The following table shows the input data (obtained from Amundsen et al., 2000). As environmental data are usually log normally distributed, median values were used for the BCF model comparison.

**Table A IV-1**. Input data for six Norwegian regions as used by Amundsen et al. (2000). Cd concentrations in soil are given in mg kg<sup>-1</sup> DW.

Region	1	2	3	4	5	6
Mean Cd	0.18	0.21	0.21	0.11	0.75	1.39
Median Cd	0.17	0.19	0.19	0.11	0.72	1.13
pH mean	5.4	4.9	4.9	5.2	5	5.3
pH median	5.4	4.9	4.9	5.2	4.9	5.2
SOM mean (%)	4.7	8.1	10.1	7.5	6.8	5.2
SOM median (%)	4	7.2	9.5	7	5.9	4.4
precipitation	680	1182	1225	824	1207	1092
(L/m2/yr)						
Drainage	300	700	900	600	1000	900
(ml/cm2/yr)*10						
net	380	482	325	224	207	192

Only a few measured data for Cd uptake into Norwegian crops are provided in Amundsen et al. (2000). These data are already the average of several studies. Additional values are given in Smolders (2001).

The EU data were derived as national medians (EU 2007). The Danish data were derived from soils with a Cd level ranging from <0.5 mg/kg DW to >2 mg/kd DW (Warming et al. 2015).

**Table A IV-2a**. Measured Cd concentrations in crops (μg kg<sup>-1</sup> DW) (Amundsen et al., 2000), ECB (2007) and (Warming et al. 2015).

	region 1 to 5	region 6 alum shale	EU	Copenhagen 2013
wheat	47	77	38 to 70	
barley	14	220		
oat	22	no data		
potato	1	10	30 to 51	32 to 83
grass	31	51		
carrot	270			90 to 206

**Table A IV-2b**. Table copied from ECB (2007); measured Cd concentrations in wheat grain and potato tuber ( $\mu$ g kg<sup>-1</sup> DW).

Table 3.182 The Cd content in selected agricultural crops and the estimated annual crop offtake

Crop	Crop Cd <sup>†</sup> μg kg <sup>-1</sup>	Comment	Typical yield (tonnes)	Crop offtake g ha <sup>-1</sup>
wheat grain	38 (M)	UK <sup>2</sup> , n=393	7.7	0.29
	58 (M)	France <sup>3</sup> , n=16	6.5	0.38
	70 (M)	The Netherlands <sup>4</sup> , n=84	8.8	0.62
	40-69 (M)	Sweden <sup>1</sup> , n=354, averages of three data sets	6.0	0.240.41
	56 (M)	Germany <sup>5</sup> , n=886	6.9	0.39
potato tuber	51 (M)	Sweden <sup>1</sup> , n=69	30	1.53
	30 (M)	The Netherlands <sup>4</sup> , n=94	35	1.05

<sup>†</sup> Mean;

) Weigert et al., 1984.

The following tables show the calculated results, using the input data given in the table, and the three methods described above.

**Table A IV-3a**. Calculated concentrations in crops, using the ERM method suggested by Amundsen et al. (2000) (method i), given as  $\mu g \ kg^{-1}$  DW.

Region	1	2	3	4	5	6
Amundsen, cereals (µg/kg DW)	42.4	49.5	49.5	38.4	102.5	140.4
Amundsen, potatoes (µg/kg DW)	53.5	66.3	66.3	56.0	87.0	95.8
Amundsen, carrots (µg/kg DW)	398	480.5	480.5	431	480.5	431

m Median;

Source;

Eriksson et al., 1996;

Chaudri et al., 1995;

Mench et al., 1997;

Wiersma et al., 1986;

**Table A IV-3b**. Calculated concentrations in crops, using the transfer factors (TF) from Smolders (2013) (method ii), given as  $\mu g \ kg^{-1}$  FW.

Region	1	2	3	4	5	6
Smolders, wheat (µg/kg FW)	23.8	26.6	26.6	15.4	100.8	158.2
Smolders, potato (µg/kg FW)	10.2	11.4	11.4	6.6	43.2	67.8
DW with 15% (grain)	28	31	31	18	119	186
DW with 70% (potato)	34	38	38	22	144	226

**Table A IV-3c**. Calculated concentrations in plants using the mass balance model (method iii), note given both as  $\mu g \ kg^{-1} \ FW$  and  $\mu g \ kg^{-1} \ DW$ .

Kd (L/kg)	194.8	172.3	214.5	239.7	165.6	186.8
C soil solution (median) ug/L	0.87	1.10	0.89	0.46	4.35	6.05
Uptake, Q * Cw, μg/kg FW	331.6	531.5	287.9	102.8	900.2	1161.6
Plus deposition µg/m2	332.5	533.0	289.1	103.2	900.4	1162.2
Plants, μg/kg FW	33.2	53.3	28.9	10.3	90.0	116.2
μg/kg DW with 15% (grain)	39	63	34	12	106	137
μg/kg DW with 70% (potato, carrots)	111	178	96	34	300	387

Concentrations of heavy metals in crops are typically measured as  $\mu g/kg$  DW, whereas consumption data are given in FW. For recalculation of DW to FW, the following equation can be used:

C(DW) = C(FW) / (1-water content)

DW concentrations can be up to 20 time higher (for example, lettuce has a water content of 95%). Typical water contents are given in **Table A IV-4**. The DW values used for calculation are the same as used in earlier calculations and are very close to values given in an ordinary nutrition table (Elmadfa et al., 1991).

**Table A IV-4.** Water content of crops. Nutrition table is from Elmadfa et al., (1991).

Crop	% water content used in calculation	% water content in nutrition table	Comment
Wheat	15	15	dried grain
Barley	15	11.7	dried grain
Oats	15	13.0	dried grain
Potato	70	77.8	raw unpeeled
Carrot	70	88.2	raw unpeeled

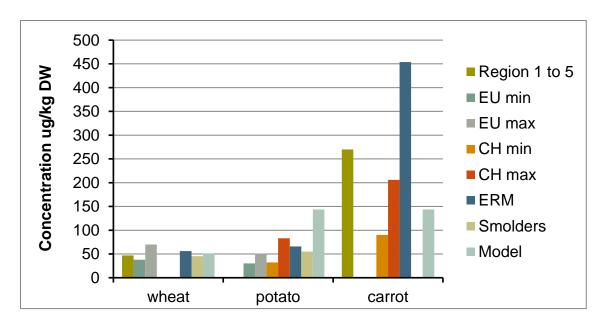
#### <u>Calculated results compared with measured data</u>

The three different methods – TF (Smolders, 2013), empirical regressions to soil properties (Amundsen et al., 2000; ERM, 2000), and the mass flux-based model (adapted from Legind et al., 2011) were compared for the six Norwegian regions, and the main crops (cereals, potatoes). Figure A IV-1 shows the values displayed in Tables A IV-2 a-b and A IV-3 a-c, i.e., the average measured concentration from regions 1 to 5 and additional measured data from the EU risk assessment report on Cd (EU 2007) and from Copenhagen (Warming et al., 2015), compared with the predicted values.

For wheat (grain), measured data from the Norwegian regions 1 to 5 and from Europe are similar and agree well with the outcome of all three prediction methods; losest is the TF-derived Smolders' value.

The measured average content of Cd in Norwegian potatoes was only 1  $\mu$ g/kg (Table A IV-2 a-b) and hardly visible on the figure. We suspect that this measurement from Norway is an outlier. ECB (2007) gives a national median value for Sweden of 51  $\mu$ g/kg DW. Furthermore, the measured values from Copenhagen (32 to 83  $\mu$ g/kg) are orders of amounts higher. These latter measurements are close to the predicted values, with the TF-value again closest.

For carrots, the average of Norwegian values is 270  $\mu$ g/kg, somewhat higher than the values from Copenhagen (90 to 206  $\mu$ g/kg). The ERM regression overestimates, while the model prediction is correct. Smolders did not derive a TF-value for carrots.



**Fig. A IV-1**. Comparison of measured concentration of Cd in crops and predicted concentrations ( $\mu$ g/kg DW). Regions 1 to 5 are the average Cd concentrations in crops grown in region 1 to 5 (Table A IV-2 a-b); EU is range given in ECB (2007) and CH are maximum and minimum values from Copenhagen (Warming et al., 2015). Predicted values are ERM, Smolders and Model, the other values are measured.

The highest Cd concentration are found in region 6, alum shale. The measured concentration in wheat is 77  $\mu$ g/kg; the predictions are 140  $\mu$ g/kg by ERM regression, 186  $\mu$ g/kg by Smolders TF, and 137  $\mu$ g/kg by the dynamic model (Table A IV-4). The same pattern is seen for potatoes and carrots: all methods show severe overprediction. It has previously been observed that there is a poor correlation between Cd contents in soil and plants (Mench et al., 1997; Warming et al., 2015), and high Cd concentrations in soil are not necessarily accompanied by high concentrations in crops, although this was predicted by all three methods.

**Table IV-5**. Measured and predicted concentrations of Cd in crops grown on alum shale (region 6), given as  $\mu g \ kg^{-1} \ DW$ .

	Measured	ERM	Smolders	Model
wheat	77	140	186	137
potato	10	96	226	387
carrot	51	431	Not included	387

In summary, TF-values, the simplest method to use, provided results that were the closest to those measured earlier, except for crops grown on alum shale. Interestingly, a similar result was found in a model comparison by Legind and Trapp (2010). Furthermore, in the study of Novotná et al. (2015), the most complex approach predicted values for uptake of Cd that were furthest from the actual values. **It was therefore decided to use the TF approach** 

**to predict the uptake of Cd into crops in this risk assessment.** The disadvantage is that Smolders only provided TF-values for wheat and potatoes. Missing TF-values were therefore taken from Legind and Trapp (2010). **Table A IV-6** displays all TF-values used in this study, and where they were obtained.

**Table A IV-6**. Transfer factors, TF, for uptake of Cd into plants (unit:  $\mu g \ kg^{-1} \ DW \ plant : \mu g \ kg^{-1} \ DW \ soil)$ .

Plant crop	TF	Reference
Leafy vegetables	0.36	US EPA, cited in Legind and Trapp (2010)
Carrots	0.25	Legind and Trapp (2010), water content 0.88
Potatoes	0.27	Smolders (2013, water content 0.778
Cereals	0.165	Smolders (2013), water content 0.15
Grass	0.36	US EPA, cited in Legind and Trapp (2010)

#### **Conclusion**

For cereals, all three methods gave rather similar results, which are within the range of reported data. Therefore, each of the methods would be suitable for the purpose of the Cd mass balance and risk assessment study. For potatoes, another relevant crop, all three methods overestimated the measured mean of Cd in potato tubers. The overestimate is for crops grown on alum shale. However, data from other European countries are very close to the predictions. Generally, measured values of Cd in crops should be preferentially used, when available.

# Appendix V

# **No Observed Effect Concentrations (NOECs) for terrestrial plants**

**Table A V-1.** NOECs (mg Cd kg<sup>-1</sup> DW soil) used for calculation of HC5 for terrestrial plants.

Species	Soil	pН	% OC	% clay	Endpoint	NOEC	Reference
Picea sitchensis	peaty gley	3.3	45	// City	root length	1.8	Burton et al., 1984
Triticum aestivum	phaeosem	6.9	1.3	21	shoot dry weight	7.1	Reber, 1989
Triticum aestivum	neutral sande hortisol	7.0	1.4	3	shoot dry weight	29	Reber, 1989
Glycine max	silt loam	7.9			shoot dry weight	10	Miller et al., 1976
Glycine max	silt loam	6.0			shoot dry weight	10	Miller et al., 1976
Glycine max	silt loam	6.5			shoot dry weight	5	Miller et al., 1976
Glycine max	clay loam	6.1			shoot dry weight	10	Miller et al., 1976
Raphanus sativus	loamy sand	5.4			shoot dry weight	10	Khan & Frankland 1983
Lactuca sativa	soil	3.9	1.2		shoot dry weight	2	Jasiewicz 1994
Lactuca sativa	humic sand	5.1	2.2		shoot dry weight	32	Adema & Henzen, 1989
Lactuca sativa	loam	7.5	0.8		shoot dry weight	3.2	Adema & Henzen, 1989
Lycosperisicon esculentum	loam	7.5	0.8		shoot dry weight	32	Adema & Henzen, 1989
Avena sativa	humic sand	5.1	2.2		shoot dry weight	10	Adema & Henzen, 1989
Avena sativa	loamy sand	7.5	0.8		shoot dry weight	10	Adema & Henzen, 1989
Phaseolus vulgaris	silt-loam with 1% clean sludge	7.5			bean dry weight	20	Bingham et al., 1975
Glycine max	silt-loam with 1% clean sludge	7.5			bean dry weight	2.5	Bingham et al., 1975
Triticum aestivum	silt-loam with 1% clean sludge	7.5			grain weight	20	Bingham et al., 1975
Zea mays	silt-loam with 1% clean sludge	7.5			kernel weight	10	Bingham et al., 1975
Lycosperisicon esculentum	silt-loam with 1% clean sludge	7.5			ripe fruit weight	80	Bingham et al., 1975
Cucurbita pepo	silt-loam with 1% clean sludge	7.5			fruit weight	80	Bingham et al., 1975
Lactuca sativa	silt-loam with 1% clean sludge	7.5			head weight	5	Bingham et al., 1975
Lepidium sativum	silt-loam with 1% clean sludge	7.5			shoot weight	5	Bingham et al., 1975
Brassica rapa	silt-loam with 1% clean sludge	7.5			tuber weight	10	Bingham et al., 1975
Raphanus sativus	silt-loam with 1% clean sludge	7.5			tuber weight	40	Bingham et al., 1975
Daucus carota	silt-loam with 1% clean sludge	7.5			tuber weight	10	Bingham et al., 1975
Lactuca sativa	surface soil with 1% clean sludge	4.8	2.6	8.3	shoot dry weight	40	Mahler et al., 1978
Beta vulgaris	surface soil with 1% clean sludge	4.8	2.6	8.3	shoot dry weight	20	Mahler et al., 1978
Lactuca sativa	surface soil with 1% clean sludge	5.0	3.3	14.6	shoot dry weight	40	Mahler et al., 1978
Beta vulgaris	surface soil with 1% clean sludge	5.0	3.3	14.6	shoot dry weight	20	Mahler et al., 1978
Lactuca sativa	surface soil with 1% clean sludge	5.3	0.9	8.9	shoot dry weight	10	Mahler et al., 1978
Beta vulgaris	surface soil with 1% clean sludge	5.3	0.9	8.9	shoot dry weight	40	Mahler et al., 1978
Lactuca sativa	surface soil with 1% clean sludge	5.7	3	37.5	shoot dry weight	20	Mahler et al., 1978
Beta vulgaris	surface soil with 1% clean sludge	5.7	3	37.5	shoot dry weight	40	Mahler et al., 1978
Lactuca sativa	surface soil with 1% clean sludge	7.4	1.4	18.7	shoot dry weight	20	Mahler et al., 1978
Beta vulgaris	surface soil with 1% clean sludge	7.4	1.4	18.7	shoot dry weight	40	Mahler et al., 1978
Lactuca sativa	surface soil with 1% clean sludge	7.5	0.6	4.4	shoot dry weight	2.5	Mahler et al., 1978
Beta vulgaris	surface soil with 1% clean sludge	7.5	0.6	4.4	shoot dry weight	20	Mahler et al., 1978
Lactuca sativa	surface soil with 1% clean sludge	7.7	0.9	40.6	shoot dry weight	5	Mahler et al., 1978
Beta vulgaris	surface soil with 1% clean sludge	7.7	0.9	40.6	shoot dry weight	40	Mahler et al., 1978
Lactuca sativa	surface soil with 1% clean sludge	7.8	0.7	15.2	shoot dry weight	10	Mahler et al., 1978
Beta vulgaris	surface soil with 1% clean sludge	7.8	0.7	15.2	shoot dry weight	80	Mahler et al., 1978
Triticum aestivum	entisol	4.1	1.1	4.9	shoot, root, germination	40	De Oliveira et al., 2016
Triticum aestivum	entisol	6.4	1.1	4.9	shoot, root	80	De Oliveira et al., 2016
Phaseolus vulgaris	entisol	4.1	1.1	4.9	shoot, root	10	De Oliveira et al., 2016
Phaseolus vulgaris	entisol	6.4	1.1	4.9	shoot, root	40	De Oliveira et al., 2016
Zea mays	entisol	4.1	1.1	4.9	root	40	De Oliveira et al., 2016
Zea mays	entisol	6.4	1.1	4.9	shoot	40	De Oliveira et al., 2016
Daucus carota	entisol	4.1	1.1	4.9	shoot, root, germination	10	De Oliveira et al., 2016
Daucus carota	entisol	6.4	1.1	4.9	germination	40	De Oliveira et al., 2016
Lactuca sativa	entisol	4.1	1.1	4.9	shoot, root	5*	De Oliveira et al., 2016
Lactuca sativa	entisol	6.4	1.1	4.9	shoot	10	De Oliveira et al., 2016
Beta vulgaris	entisol	4.1	1.1	4.9	shoot	5*	De Oliveira et al., 2016
Beta vulgaris	entisol	6.4	1.1	4.9	shoot	40	De Oliveira et al., 2016
Avena strigosa	entisol	4.1	1.1	4.9	germination	10	De Oliveira et al., 2016
Avena strigosa	entisol	6.4	1.1	4.9	shoot, root	80	De Oliveira et al., 2016
Oryza sativa	entisol	4.1	1.1	4.9	shoot, root, germination	40	De Oliveira et al., 2016
Oryza sativa	entisol	6.4	1.1	4.9	shoot, root, germination	160	De Oliveira et al., 2016
Avena sativa	Natural loamy soil	6.6	1.5	26	growth (biomass)	6.25	da Rosa Corrêa et al. 2006
Lactuca satvia	Natural loamy soil	6.6	1.5	26	growth (biomass)	3.12	da Rosa Corrêa et al. 2006
Brassica acmprestis	Natural loamy soil	6.6	1.5	26	growth (biomass)	25	da Rosa Corrêa et al. 2006
	* LOEC/2						

# Appendix VI

## **Soil properties**

The soil properties/parameters pH and mould% from NIBIO Soil database was collected and summarised in Table A VI-1-2. Number of samples, minimum, maximum and mean for the four municipality cases and the six regions in Norway, is shown.

**Table A VI-1.** Mean, minimum, and maximum  $pH(H_2O)$  in agricultural soils in different municipalities, regions, and for the whole country (NIBIO Soil database). pH used for risk assessment is adjusted for  $pH(CaCl_2)$  (Eq.1).

Municipality	Minimum	Maximum	Mean	No. of samples
ÅS	3.2	8.7	6.1	8829
STANGE	3.5	8.4	6.1	7703
TIME	2.6	7.8	5.8	4389
MELHUS	4.3	8.4	6.2	2251

Region	Minimum	Maximum	Mean	No. of samples
Northern Norway	1.5	9.9	5.9	52425
Southeastern Norway	1	13.4	6.1	531568
Southern Norway	2.5	8.7	5.7	25849
Trøndelag (Mid Norway)	1	11.1	6.0	120491
Western Norway	0.9	9.5	5.8	201149
Country	0.9	13.4	6.0	931482

**Table A VI-2.** Mean, minimum, and maximum of mould% (same as SOM%) in agricultural soils for different municipalities, regions, and for the whole country (NIBIO Soil database).

Municipality	Minimum	Maximum	Mean	No. of samples
ÅS	0.7	85.2	5.7	8799
STANGE	0.51	97.1	6.7	7503
TIME	1	31.35	4.1	4383
MELHUS	0.6	92	6.3	2205

Region	Minimum	Maximum	Mean	No. of samples
Northern Norway	0.55	98.8	9.9	47939
Southeastern Norway	0.51	99	6.0	517982
Southern Norway	0.57	98.8	11.0	23636
Trøndelag (Mid Norway)	0.58	98.8	8.0	114801
Western Norway	0.59	98.8	10.6	187627
Country	0.51	99	7.6	891985

**Table A VI-3.** Mean, minimum and maximum of adjusted volum weight (Soil dry density) in agricultural soils for different municipalities, regions and for the whole country (NIBIO Soil database).

Municipality	Minimum	Maximum	Mean	No of samples
ÅS	0.24	14	1.3	8881
STANGE	0.07	2	1.3	7712
TIME	0.51	1.09	0.8	11
MELHUS	0.03	1.92	1.2	2272

Region	Minimum	Maximum	Mean	No of samples
Northern Norway	0.03	2	1.1	52495
Southeastern Norway	0.03	14	1.2	531797
Southern Norway	0.05	2	1.0	25927
Trøndelag (Mid Norway)	0.03	18.7	1.1	120310
Western Norway	0.01	5.9	1.0	179024
Country	0.01	18.7	1.2	909553