# Greenhouse gas emissions from cultivated and abandoned organic croplands in Finland

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Human activities have enhanced the emissions of carbon dioxide ( $\rm CO_2$ ) and nitrous oxide ( $\rm N_2O$ ) from organic soils. Drained peat soils may turn into a source of  $\rm CO_2$  and  $\rm N_2O$ , whereas  $\rm CH_4$  emission may generally decrease after drainage. Agricultural use, including ploughing, fertilization and irrigation, further enhances the decomposition of peat, and  $\rm CO_2$  and  $\rm N_2O$  emissions. Cultivated organic soils are therefore significant sources of  $\rm CO_2$  and  $\rm N_2O$ . In this paper we report greenhouse gas emissions from cultivated organic soils, and from abandoned organic agricultural soils, i.e. from organic soils where cultivation practices have ceased. We found that  $\rm CO_2$  and  $\rm N_2O$  emissions from abandoned organic cropland soils do not generally decrease with time after agricultural practices have ceased, whereas  $\rm CH_4$  fluxes from the atmosphere into the soil may gradually increase after leaving cultivation.

#### Introduction

Of the original peatland area of 10.4 million ha about 0.7 million ha have been cleared and drained for agriculture in Finland. About half of these soils have been abandoned, afforested or have become mineral soils (Myllys 1996). Of the organic soils, including peat and mull soils, 300 000 ha are still in agricultural use (Myllys and Sinkkonen 2004).

In agriculture, a low water table level, repeated ploughing of the soil, fertilization, liming (increase in pH) and mineral soil addi-

tion enhance the decomposition of organic soil and associated carbon dioxide (CO<sub>2</sub>) production. In organic agricultural soils, the decomposition of organic matter is higher than the uptake of CO<sub>2</sub> by plants and therefore there is a net loss of CO<sub>2</sub> from the drained organic agricultural soils (Kasimir-Klemedtsson *et al.* 1997, Maljanen *et al.* 2001, 2004, Lohila *et al.* 2004).

Nitrous oxide (N<sub>2</sub>O) is 296 times as effective greenhouse gas as CO<sub>2</sub> in a 100-year time horizon (Houghton *et al.* 2001). Agricultural soils are responsible for most of the global N<sub>2</sub>O emissions from soils (Kroeze *et al.* 1999). Drainage



**Fig. 1.** The study sites in Finland. Cultivated organic agricultural soils were measured in: 1 = Jokioinen (Lohila *et al.* 2004, Regina *et al.* 2004; K. Regina *et al.* unpubl. data), 2 = Liperi (Maljanen *et al.* 2001, 2003a, 2003b, 2003c), 3 = Ilomantsi (Nykänen *et al.* 1995), 4 = Rovaniemi (Regina *et al.* 2004; K. Regina *et al.* unpubl. data), 5a = Kannus (Maljanen *et al.* 2004), and 5b = abandoned organic agricultural soils in Kannus (previously unpublished).

increases emissions of  $N_2O$  from organic soils (Martikainen *et al.* 1993) and drained organic soils have special importance in the atmospheric  $N_2O$  load. As much as 25% (annually 4 Tg) of the  $N_2O$  emissions in Finland may originate from organic agricultural soils (Kasimir-Klemedtsson *et al.* 1997), although the coverage of these soils is only 13.6% of the total agricultural land in the country (Myllys and Sinkkonen 2004).

N<sub>2</sub>O is produced in soil microbial activi-

ties, with nitrification and denitrification as the key processes. Temperature, soil moisture, soil pH, water table level, vegetation, ploughing, compaction of soil by machinery and availability of organic carbon affect the N<sub>2</sub>O emissions (Freney 1997, Augustin et al. 1998). In organic soils the availability of mineral nitrogen and organic microbial substrates are high, which favour the microbial processes associated to the N<sub>2</sub>O production. However, the environmental factors controlling the N<sub>2</sub>O fluxes form a complex network (Groffmann et al. 2000) and the N<sub>2</sub>O fluxes are difficult to predict. In the boreal region the winter emissions of N<sub>2</sub>O contribute significantly to the annual N<sub>2</sub>O budget, but they are still poorly quantified. Winter and early spring emissions may account for more than half of the annual N<sub>2</sub>O emissions in the boreal region (e.g. Maljanen et al. 2003b, Regina et al. 2004).

Methane (CH<sub>4</sub>) is 23 times as powerful a greenhouse gas as CO<sub>2</sub> in a 100-year time horizon (Houghton et al. 2001). CH<sub>4</sub> from agriculture originates mainly from the digestion of ruminants, manure management (Rossi et al. 2001) and from waterlogged soils, like rice paddies, where anaerobic conditions dominate (Le Mer and Roger 2001). The well drained agricultural soils are generally sinks for atmospheric CH<sub>4</sub> and for CH<sub>4</sub> produced in the deeper soil layers because more CH<sub>4</sub> is consumed (oxidized) in the aerobic soil by methanotrophic bacteria than is produced by methanogens (Le Mer and Roger 2001). Drained organic soils used for agriculture can be either small sinks or sources for CH<sub>4</sub> (Nykänen et al. 1995, Augustin et al. 1998, Maljanen et al. 2003a) depending on the water table level and weather conditions. Nitrogen fertilization, liming and changes in the physical structure of soil caused by tilling or machinery may affect the CH<sub>4</sub> oxidation in agricultural soils (Mosier et al. 1991, 1997, Powlson et al. 1997, Hütsch 2001). Soil moisture and temperature influence CH<sub>4</sub> production and consumption in soils and thus account for the seasonal variation in the CH<sub>4</sub> fluxes (Hütsch et al. 1994).

After the cultivation practices have ceased, gradual secondary vegetation succession starts. Grasses and herbs still dominate in field vegetation for 10–15 years (Törmälä 1982). Open field ditches are the first habitat for the pioneer tree

species (birch, willows) (Hytönen 1999). The gradual deterioration of the ditch network leads to higher water table levels, which may increase CH<sub>4</sub> emissions. On the other hand, ending cultivation practises, e.g. ploughing and fertilization, may reduce the decomposition rate of peat and also CO<sub>2</sub> and N<sub>2</sub>O emissions.

The annual greenhouse gas emissions from Finnish organic agricultural soils in active use (grassland, cropland) were measured at five sites (Nykänen *et al.* 1995, Maljanen *et al.* 2001, 2003a, 2003b, 2004, Lohila *et al.* 2004, Regina *et al.* 2004, K. Regina *et al.* unpubl. data). In this paper we summarize the results of these studies. We also report the annual and seasonal greenhouse gas emissions from five abandoned organic agricultural soils in Finland and discuss how greenhouse gas emissions change after cultivation practices have ceased.

#### Materials and methods

# GHG measurements from cultivated agricultural soils

Annual N<sub>2</sub>O and CH<sub>4</sub> measurements were carried out at five different sites from north to south Finland in 1991–2002 (Nykänen *et al.* 1995, Maljanen *et al.* 2003a, 2003b, 2004, Regina *et al.* 2004; K. Regina *et al.* unpubl. data; Fig. 1). N<sub>2</sub>O and CH<sub>4</sub> fluxes were measured using a chamber method (Nykänen *et al.* 1995). CO<sub>2</sub> balances were measured at two sites using a chamber

method (Maljanen *et al.* 2001, 2004) and at two sites using the eddy covariance method (Lohila *et al.* 2004). The measurements were made on soils under barley or potato and grass. Also sites with no vegetation (fallow) were studied. Some soil properties of the sites are shown in Table 1.

# GHG measurements from abandoned organic agricultural soils

Study sites

N<sub>2</sub>O and CH<sub>4</sub> fluxes were measured during the years 2003 and 2004 on five abandoned organic agricultural fields in Kannus, western Finland (63°54′N, 23°56′E, Fig. 1). There were altogether 30 gas sampling plots on these fields (6 plots at each site). These organic soils were drained and used for cultivation of grass and cereals for decades before they were abandoned 20 to 30 years ago — no fertilisation or ploughing activities have been carried out since then. The water table levels varied between 154 cm below to 5 cm above the soil surface, being on average 46 cm below the soil surface in May-October. The depth of peat varied from 0.2 to > 1 m. Organic matter content in the top soil (0-10 cm) was on average 48%. The pH range in these soils was from 4.3 to 5.9. The most common plant species at the sites were Agrostis sp., Carex sp., Cirsium palustre, Deschampsia cespitosa, Elymus repens, Epilobium angustifolium, Galium sp., Juncus filiformis and Poa pratensis.

**Table 1**. Soil properties of the cultivated and abandoned organic soils. The cultivated sites were: 1 = Jokioinen (Regina *et al.* 2004; K. Regina *et al.* unpubl. data), 2 = Liperi (Maljanen *et al.* 2001, 2003a, 2003b), 3 = Ilomantsi (Nykänen *et al.* 1995), 4 = Rovaniemi (Regina *et al.* 2004; K. Regina *et al.* unpubl. data), 5a = Kannus, two subsites with different peat depths (Maljanen *et al.* 2004), 5b = Kannus, abandoned organic agricultural soils in Kannus (previously unpublished) (*see* Fig. 1).

Site	C/N	Bulk density (g cm <sup>-3</sup> )	pH (H <sub>2</sub> O)	Peat depth (m)	Since drainage (years)
1	21	0.49-0.51	5.8	nd	100
2	16	0.33	6.0	0.2	40
3	19	nd	5.3	1.4	60
4	18	0.24-0.29	5.6	nd	50
5a	31-32	0.32-0.50	4.8	0.3 to 0.7	nd
5b	16–19	0.33-0.47	4.3–5.9	0.2  to > 1.0	50–100

## CO, balance measurements

CO<sub>2</sub> exchange between the soil-vegetation system and the atmosphere was measured using a chamber method in 2003. The net ecosystem CO<sub>2</sub> exchange (NEE) was measured with a transparent chamber (Alm et al. 1997). This climatecontrolled chamber ( $60 \times 60$  cm, height 30 cm) was placed on a collar ( $56 \times 56$  cm). The collar had a groove in the upper edge which was filled with water to ensure a gas tight seal. A portable infrared gas analyzer (EGM-4 Environmental Gas Monitor for CO2, PP Systems, UK) monitored the change in the CO<sub>2</sub> concentration in the chamber during the chamber closure period of 0 to 180 seconds. Simultaneously the light intensity (PAR) and air temperature  $(T_a)$  inside the chamber were recorded. The measurements were made in full light and then in reduced light (ca. 70%) under mosquito netting. After that the gross respiration rate  $(R_{TOT})$  of the plant-soil system was measured in the same way by covering the collar with an opaque cover.

 $\mathrm{CO_2}$  flux rates, NEE (net ecosystem exchange; measurement with transparent chamber) and  $R_{\mathrm{TOT}}$  (total respiration; measurement with an opaque chamber), were calculated from the linear increase or decrease in the  $\mathrm{CO_2}$  concentration in the headspace of the chamber.  $\mathrm{CO_2}$  uptake from the atmosphere is designated with a positive, and release of  $\mathrm{CO_2}$  to the atmosphere with a negative sign. NEE can be positive or negative while  $R_{\mathrm{TOT}}$  is always negative. An estimate of gross photosynthesis ( $P_{\mathrm{G}}$ ) was calculated as the sum of measured  $\mathrm{CO_2}$  fluxes in light (NEE) and dark ( $R_{\mathrm{TOT}}$ ) (Alm *et al.* 1997),

$$NEE = P_G - R_{TOT}.$$
 (1)

These measurements were made from each collar on the abandoned fields weekly or biweekly during the growing season.

For calculation of the diurnal cycles of NEE, the values for  $P_{\rm G}$  and  $R_{\rm TOT}$  are needed for every hour. Statistical response functions were constructed separately for each site during the growing season 2003, in order to reconstruct diurnal cycles of  $P_{\rm G}$  and  $R_{\rm TOT}$  on the basis of climatic data (e.g. Alm *et al.* 1997). The dependence of  $P_{\rm G}$  on irradiation was modelled using a form of

rectangular hyperbola, where the single variable I is the photosynthetically active radiation (E, m<sup>-2</sup> s<sup>-1</sup>), and Q and k are asymptotic maximum and half saturation constant, respectively. As  $P_{\rm G}$  depends on the rate of soil  ${\rm CO}_2$  release into the chamber, variables explaining this, LAI (leaf area index),  $T_{\rm 5}$  (soil temperature at the depth of 5 cm, °C) and WT (water table level) were included. Parameters (b) for the hyperbolic functions were estimated with a non-linear Levenberg-Marquard technique using the estimated values for  $P_{\rm G}$  and measured values for I.

$$\begin{split} P_{\rm G} &= \left[ (Q \times I)/(k+I) \right] + b_0 + b_1 \times T_5 \\ &+ b_2 \times \text{LAI} + b_3 \times \text{WT} \end{split} \tag{2}$$

The response functions for  $\ln R_{\rm TOT}$  were linear models with two explanatory variables, soil surface temperature ( $T_{\rm 5}$ ) and soil water table level (WT). Parameter values were estimated using linear regression (SPSS).

$$\ln R_{\text{TOT}} = b_0 + b_1 T_5 + b_2 \text{WT}$$
 (3)

The diurnal cycles of  $P_{\rm G}$  were then reconstructed using the response functions and continuous data for LAI,  $T_{\rm S}$ , WT and I, and those of  $R_{\rm TOT}$  using continuous data for  $T_{\rm S}$  and WT. The net CO<sub>2</sub> emission for the growing season was calculated from the hourly values of NEE. Outside the growing season, the net CO<sub>2</sub> release from soil was measured biweekly similarly as N<sub>2</sub>O and CH<sub>4</sub> (see N<sub>2</sub>O and CH<sub>4</sub> flux measurements). The concentration of CO<sub>2</sub> in the gas samples was analysed with a gas chromatograph used for N<sub>2</sub>O and CH<sub>4</sub> analysis equipped with a TC (thermal conductivity) detector.

# N<sub>2</sub>O and CH<sub>4</sub> flux measurements

Gas flux measurements were made throughout two years. During the snow free periods, fluxes of CH<sub>4</sub> and N<sub>2</sub>O were measured every second or third week with a static chamber method (Nykänen *et al.* 1995). Permanent aluminium collars (58 × 58 cm) and an aluminium chamber (60 × 60 cm, height 30 cm, equipped with a fan) were used. The collar had a groove in the upper edge which was filled with water to ensure a

gas-tight seal. Gas samples (40 ml) were drawn from the headspace of the chambers with 60ml polypropylene syringes (Terumo or Becton Dickinson) equipped with a three-way stopcock 5, 10, 15 and 25 minutes after the chambers were installed. Gas concentrations were analysed within 24 hours from sampling with gas chromatographs (Shimadzu GC-14B or Hewlett-Packard 5890) equipped with flame ionisation (FI) and electron capture (EC) detectors (Nykänen et al. 1995, Maljanen *et al.* 2003a, 2003b). The fluxes were calculated from the linear change in the gas concentrations in the headspace of the chamber. During winter, the gas fluxes were determined with a gas gradient technique (Maljanen et al. 2003c). Gas samples for concentration analyses of N<sub>2</sub>O and CH<sub>4</sub> were drawn from the snow pack using a stainless steel probe 3 mm in diam. Gas fluxes from soil through the snow pack were calculated using Fick's law (Sommerfeld et al. 1993).

# Statistical analyses

The correlations between annual emissions and environmental variables were calculated using Spearman's rank correlation (SPSS 11.5.1. SPSS Inc.).

### Results and discussion

The cultivated organic cropland soils were all net sources of CO<sub>2</sub> (Table 2). The CO<sub>2</sub> losses measured with the chamber method from soil under grass varied from 79 to 750 g CO<sub>2</sub>-C m<sup>-2</sup> and CO<sub>2</sub> losses from soils under barley were from 210 to 830 g CO<sub>2</sub>-C m<sup>-2</sup>. The net CO<sub>2</sub> emissions from the fallow soils (without vegetation) had similar CO<sub>2</sub> net loss (from 690 to 1100 g CO<sub>2</sub>-C m<sup>-2</sup>), as that at the barley fields (Maljanen et al. 2001, 2004). The EC measurements showed lower CO, release than the chamber measurements (Lohila et al. 2004). The CO<sub>2</sub> balance is sensitive to climatic conditions and therefore this difference can be partly explained by the variation in the environmental factors controlling CO, flux because the EC and chamber measurements were done in different years. However, the emissions are close to those, 400–550 g CO<sub>2</sub>-C m<sup>-2</sup>, estimated earlier for boreal organic agricultural soils (Kasimir-Klemedtsson et al. 1997).

The abandoned agricultural soils were annually small net sinks (up to 90 g  $\rm CO_2\text{-}C~m^{-2}$ ) or sources of  $\rm CO_2$  (highest net emission 900 g  $\rm CO_2\text{-}C~m^{-2}$ ). There was some net  $\rm CO_2$  uptake during the growing season, but outside that period all abandoned agricultural soils were net sources of  $\rm CO_2$ . The mean annual net  $\rm CO_2$  emis-

**Table 2**. Annual fluxes of greenhouse gases from cultivated organic soils in Finland. Minus sign indicates uptake of the gas.  $CO_2$ -C,  $CH_4$ -C and  $N_2$ O-N emissions in g m<sup>-2</sup>.

Crop	Site	CO <sub>2</sub> -C	CH <sub>4</sub> -C	$N_2O-N$	References
Barley	1	830	-0.13-0.06	0.54–1.13	Maljanen <i>et al.</i> 2004
Barley	2	210	-0.01-0.04	0.62-2.41	Lohila <i>et al.</i> 2004, Regina <i>et al.</i> 2004, K. Regina <i>et al.</i> unpubl. data.
Barley	4	nd	-0.02-0.38	0.73-1.88	Regina et al. 2004, 2006
Barley	5a	400	-0.37-0.01	0.83-0.84	Maljanen et al. 2001, 2003a, 2003b
Grass	1	330-460	-0.07-0.18	0.17-0.38	Maljanen et al. 2004
Grass	2	80	-0.05-0.01	0.50-0.99	Lohila <i>et al.</i> 2004, Regina <i>et al.</i> 2004, K. Regina <i>et al.</i> unpubl. data.
Grass	3	nd	0.27-0.68	0.26-0.53	Regina et al. 2004, K. Regina et al. unpubl. data
Grass	4	750	-0.08	1.10	Maljanen et al. 2001, 2003a, 2003b
Grass	5a	nd	0.10-0.20	0.78-0.93	Nykänen <i>et al.</i> 1995
Potato	1	nd	nd	1.04	Regina et al. 2004
Fallow	1	690-790	-0.14-0.01	0.40-3.70	Maljanen et al. 2004
Fallow	2	nd	0.03-0.01	1.34-3.70	Regina et al. 2004
Fallow	4	nd	0.04-3.00	0.38-0.50	Regina et al. 2004, K. Regina et al. unpubl. data
Fallow	5a	880-1100	-0.26-0.13	0.65-0.71	Maljanen et al. 2001, 2003a, 2003b

sion, 324 g CO<sub>2</sub>-C m<sup>-2</sup>, is close to the net CO<sub>2</sub> emissions from cultivated croplands. It seems that abandoned organic agricultural soils do not generally turn into CO<sub>2</sub> sinks after the agricultural practices have ceased, although their net CO<sub>2</sub> emissions may be slightly reduced (Table 3). Fallow soils, without vegetation, had almost similar CO<sub>2</sub> net loss as that at the barley fields.

Cultivated organic soils were either sinks or sources of atmospheric CH<sub>4</sub> (Table 2) depending on the ground water level in the site. The fallow soils without plants had a lower CH uptake rate than the cultivated soils. Depending on the weather conditions they either emitted CH<sub>4</sub> or were small sinks. However, there was no correlation between the mean annual CH<sub>4</sub> flux and the mean WT, soil pH, C, N or C:N ratio of soil. Annually all the abandoned cropland soils were net sinks for atmospheric CH<sub>4</sub> (Table 3), even with some periods with low CH<sub>4</sub> emissions. Despite the deterioration of the ditch network after 20-30 years of abandonment, the water table level remained considerably deep. The mean soil CH, uptake rate was even higher at the abandoned than at the cultivated sites. This indicates that with deep water-table levels the ability of soils to withdraw CH<sub>4</sub> from the atmosphere may gradually increase after leaving cultivation. Both the croplands and abandoned soils also showed small CH<sub>4</sub> uptake during winter.

The mean N<sub>2</sub>O emissions from croplands

under barley were higher than those from croplands under grass (Table 3). The  $\rm N_2O$  emissions from fallow soils in the studies of Nykänen *et al.* (1995), Maljanen *et al.* (2003b, 2004) and Regina *et al.* (2004) were generally higher than those from vegetated fields. This may be related to higher N-availability for denitrification in the absence of plants (Silvan *et al.* 2005).

Surprisingly, the N<sub>2</sub>O emissions from the abandoned and cultivated cropland soils were similar, with emissions from the abandoned soils even higher than those from the croplands under grass (Table 3). The time since the cultivation practices ended did not correlate with the annual N<sub>2</sub>O emission rates from the abandoned croplands. Therefore, we found no evidence that the ending of cultivation activities reduces N<sub>2</sub>O emissions from croplands on organic soils. The abandoned croplands, without N-fertilization, still have a considerable N-pool bound in the organic matter. When mineralized and nitrified this nitrogen pool provides plenty of NO<sub>3</sub>- for denitrification which is probably the main source of N<sub>2</sub>O from drained organic agricultural soils (Maljanen et al. 2003b).

There was high variation in the seasonal and annual  $N_2O$  emissions and only a part of the variation could be explained by weather conditions, e.g. temperature and precipitation. Generally the annual  $N_2O$  emissions from the croplands and abandoned croplands did not correlate with the

**Table 3.** Annual and seasonal emissions from cultivated or abandoned organic agricultural soils (mean, median and standard deviation (SD) from the reported annual emission, *n* is the number of annual emissions reported). CO<sub>2</sub>-C, CH<sub>4</sub>-C and N<sub>2</sub>O-N emissions (g m<sup>-2</sup>).

		CO <sub>2</sub> -C annual	CH <sub>4</sub> -C annual	N <sub>2</sub> O-N		
				Summer	Winter	Annual
Barley	Mean	568 (n = 4)	-0.052 (n = 12)	0.457	0.651	1.108 (n = 11)
•	Median	615	-0.068	0.520	0.330	0.850
	SD	312	0.183	0.235	0.598	0.588
Grass	Mean	405 (n = 4)	0.071 (n = 12)	0.439	0.135	0.574 (n = 12)
	Median	395	-0.020	0.340	0.060	0.410
	SD	279	0.242	0.278	0.186	0.321
Fallow (no plants)	Mean	591 $(n = 6)$	0.239 (n = 10)	1.102	0.569	1.671 ( <i>n</i> = 10)
	Median	740	-0.093	0.445	0.380	0.925
	SD	378	0.974	1.252	0.781	1.389
Abandoned (mixed vegetation)	Mean	324 (n = 5)	-0.165 (n = 5)	0.439	0.383	0.822 (n = 5)
	Median	287	-0.105	0.230	0.333	0.563
	SD	250	0.135	0.759	0.204	0.884

mean WT even though there was some correlation during the growing season (e.g. Maljanen *et al.* 2003b). When the annual emissions from croplands and abandoned organic soils were plotted against the C:N ratio of the soil (0–10 cm) there was a decreasing trend in the annual N<sub>2</sub>O emissions with the increase in the C:N ratio, but the correlation was not significant. Klemedtsson *et al.* (2005) have recently reported that the C:N ratio of organic forest soil may predict the annual N<sub>2</sub>O emissions. However, in cropland soils other factors, e.g. fertilization and ploughing, may overcome the C:N dependence which reflects the availability of mineral nitrogen for microbial processes important in N<sub>2</sub>O emissions.

It is important to notice that the winter emissions (from October to May) of  $N_2O$  were from 25% (grass) up to 60% (barley) of the annual emissions. Therefore, the winter emissions have to be included in the annual estimates of  $N_2O$  emissions from agricultural soils.

When considering the atmospheric impact of these organic croplands, CO<sub>2</sub> was the most important gas. CO<sub>2</sub> was responsible on average for 78% of the total GWP (global warming potential) caused by the sum of CO<sub>2</sub>, N<sub>2</sub>O and CH<sub>4</sub> fluxes with the 100-year time horizon (Houghton *et al.* 2001). N<sub>2</sub>O was responsible for about 22% of the total GWP. The effect of CH<sub>4</sub> was insignificant, less than 1% of the total GWP.

### Conclusion

Carbon dioxide emissions from abandoned organic cropland soils do not generally decrease with time after agricultural practices have ceased — the decomposition of drained peat evidently continues after ending of cultivation activities. The N<sub>2</sub>O emissions from croplands on organic soil can also still be high after 20–30 years of abandonment. CH<sub>4</sub> fluxes from the atmosphere into the soil may gradually increase after cultivation has ceased, but CH<sub>4</sub> has minor importance in the total GWP caused by CO<sub>2</sub>, N<sub>2</sub>O and CH<sub>4</sub>.

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