



## Boreal forests can have a remarkable role in reducing greenhouse gas emissions locally: Land use-related and anthropogenic greenhouse gas emissions and sinks at the municipality level



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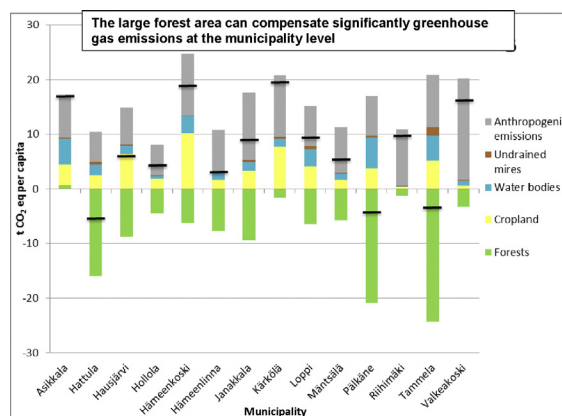
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### HIGHLIGHTS

- The significance of natural landscapes in the regional C budgets is shown.
- Boreal forests can be remarkable C sinks enabling net C sequestration in ecosystems.
- The large area of forest may compensate for all C emissions in the municipality.
- Forest management policy can be a key factor for mitigating municipal GHG emissions.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Ecosystem services have become an important concept in policy-making. Carbon (C) sequestration into ecosystems is a significant ecosystem service, whereas C losses can be considered as an ecosystem disservice. Municipalities are in a position to make decisions that affect local emissions and therefore are important when considering greenhouse gas (GHG) mitigation. Integrated estimations of fluxes at a regional level help local authorities to develop land use policies for minimising GHG emissions and maximising C sinks. In this study, the Finnish national GHG accounting system is modified and applied at the municipal level by combining emissions and sinks from agricultural land, forest areas, water bodies and mires (land use-related GHG emissions) with emissions from activities such as energy production and traffic (anthropogenic GHG emissions) into the LUONNIKAS calculation tool. The study area consists of 14 municipalities within the Vanajavesi catchment area located in Southern Finland. In these municipalities, croplands, peat extraction sites, water bodies and undrained mires are emission sources, whereas forests are large carbon sinks that turn the land use-related GHG budget negative, resulting in C sequestration into the ecosystem. The annual land use-related sink in the study area was 78 t CO<sub>2</sub> eq km<sup>-2</sup> and 2.8 t CO<sub>2</sub> eq per capita. Annual anthropogenic GHG emissions from the area amounted to 250 t CO<sub>2</sub> eq km<sup>-2</sup> and 9.2 t CO<sub>2</sub> eq per capita. Since forests are a significant carbon sink and the efficiency of

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this sink is heavily affected by forest management practices, forest management policy is a key contributing factor for mitigating municipal GHG emissions.

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## 1. Introduction

To mitigate climate change, the European Union has committed itself to a reduction in greenhouse gas (GHG) emissions of at least 20% below 1990 levels by 2020 (EC, 2008). Therefore, carbon (C) sequestration into ecosystems is an important ecosystem service. Although GHG emissions from various land use types is a well-studied environmental phenomenon (e.g. Martikainen et al., 1993, Le Mer and Roger, 2001 and Dalal and Allen, 2008), there is an urgent need for integrated estimations of fluxes at the landscape and regional levels (Buffam et al., 2011). These estimates help regional and local authorities to develop measures, land use policies and landscape management practices for the minimisation of GHG emissions. Municipalities are often in a position to make decisions that affect local emissions. In particular, municipalities are responsible for land use policies. Comprehensive municipality-level information of different GHG sources and sinks is needed for mitigating GHG emissions via planning, management and decision-making.

In this study the national GHG accounting system (Statistics Finland, 2013) is modified and applied at the municipal level. In addition, the land use-related GHG emissions and sinks are combined with the anthropogenic GHG emissions at the municipal level. We define land use-related emissions and sinks as including forest areas, croplands, water bodies such as lakes, streams and rivers, and mires. Further, emissions from peat extraction sites are included as land use-related emissions. Anthropogenic GHG emissions result from human activities such as emissions from burning fossil fuels for energy (IPCC, 2001).

In Finland there is a well-developed carbon accounting system that includes anthropogenic and land use-related GHG emissions from terrestrial ecosystems, as well as regionally representative GHG evasion estimates from aquatic ecosystems (e.g. Kortelainen et al., 2006, Bergström et al., 2007 and Juutinen et al., 2009). However, holistic studies on a municipal scale are lacking.

The aim of this study was to demonstrate the variety of GHG sources at the municipal level by applying an easy-to-use calculation tool for GHG emissions and sinks of different natural and anthropogenic sources in 14 municipalities in Southern Finland. By taking advantage of several existing free data sources, it is possible to easily calculate various GHG sinks and sources in order to support local authorities in planning and implementing more sustainable actions, strategies and management practices to reduce GHG emissions. Our hypothesis is that land use-related carbon emissions and sinks at the landscape and municipal level can be significant when compared to emissions from human activities (i.e. anthropogenic emissions).

## 2. Methods

### 2.1. Study area

The study area consists of 14 municipalities within the Vanajavesi catchment area, located in boreal zone in Southern Finland (60°40'–61°20'N, 24°10'–25°20'E). The total area is 8400 km<sup>2</sup>, comprising 13% water, including lakes and rivers. More than 70% of the land area is covered by coniferous and mixed deciduous woodland or mires. The population density ranges from 9 to 233 persons per km<sup>2</sup>, the average density in 2013 being 28 persons per km<sup>2</sup>. The map of municipalities with their total area and land use are shown in Fig. 1 and Table 1.

### 2.2. Land use-related GHG emissions and sinks

The LUONNIKAS calculation tool (Haaspuro, 2013) was used to calculate municipal-level estimates of carbon budgets for forests, cropland, mires and water bodies. LUONNIKAS calculates carbon sequestration into the ecosystem and the amount of GHG emissions for a one-year period. For the analysis we used data from year 2009 which was the most recent to achieve complete datasets. When methane (CH<sub>4</sub>) or dinitrogen monoxide (N<sub>2</sub>O) emissions were assumed to be significant, they were also added to the calculations, and the results presented as CO<sub>2</sub> equivalents.

The LUONNIKAS calculation tool consists of simple calculation methods to ensure that data needed for calculations is easily available at the municipal level. The calculation methods for GHG emissions and sinks mostly follow the methodology used in the Land Use, Land Use Change and Forestry (LULUCF) sector in Finland's national greenhouse gas inventory (Statistics Finland, 2013).

This inventory provides annual information on the national GHG emissions and removals that are reported to the United Nations Framework Convention on Climate Change (UNFCCC) and the European Commission (Statistics Finland, 2013). In the LULUCF sector, the emissions and sinks are calculated for managed land use types. It includes the carbon budgets of forest land, cropland and peat extraction sites. Inland waters and undrained mires are considered unmanaged, and therefore no emissions are estimated for those land use classes in the greenhouse gas inventory (Statistics Finland, 2013), although emissions from both these land use types can be large (Bergström et al., 2007). High nutrient concentrations have been shown to increase both CO<sub>2</sub> and CH<sub>4</sub> evasion from boreal lakes (Kortelainen et al., 2006; Juutinen et al., 2009) in agreement with recent global estimates by Lauerwald et al. (2015), which highlighted the anthropogenic drivers for CO<sub>2</sub> evasion from global river network. In this study carbon budgets of water bodies and undrained mires were calculated based on regionally representative studies, most of which were carried out in boreal Finland and Sweden (see Bastviken et al., 2004, Kortelainen et al., 2006, Saarnio et al., 2007, Juutinen et al., 2009 and Humborg et al., 2010). Inland waters also sequester terrestrially fixed carbon. However, permanent C pools in boreal lakes are minor compared to CO<sub>2</sub> evasion (Kortelainen et al., 2004; Kortelainen et al., 2006).

#### 2.2.1. Forest

The carbon budget of the forest was calculated as a sum of forest biomass increment, forest biomass removals by forest harvesting, and the carbon storage change of the forest soil. The calculation of carbon sequestration in biomass included all forests on mineral and organic soils. The forest soil carbon budget was calculated for all mineral soils and drained organic soils.

The forest biomass increment was calculated using forest areas, region-specific growth rates for different tree species and forest land types (Statistics Finland, 2013), and biomass expansion factors (BEF) specific to geographical location, organic and mineral soils, forest land types, tree species and below and above-ground biomass. The harvest removals were calculated by transforming the municipal-level data of yearly fellings in privately-owned forests into total harvest removals. The amount of C in total harvest removals was subtracted from the amount of C in biomass increment, which gives the one-year net carbon budget, i.e. C sequestered in or removed from the biomass.

The forest soil carbon budget was calculated by multiplying the area of the mineral and organic forest soil by respective emission factors from Finland's national greenhouse gas inventory (Statistics Finland,

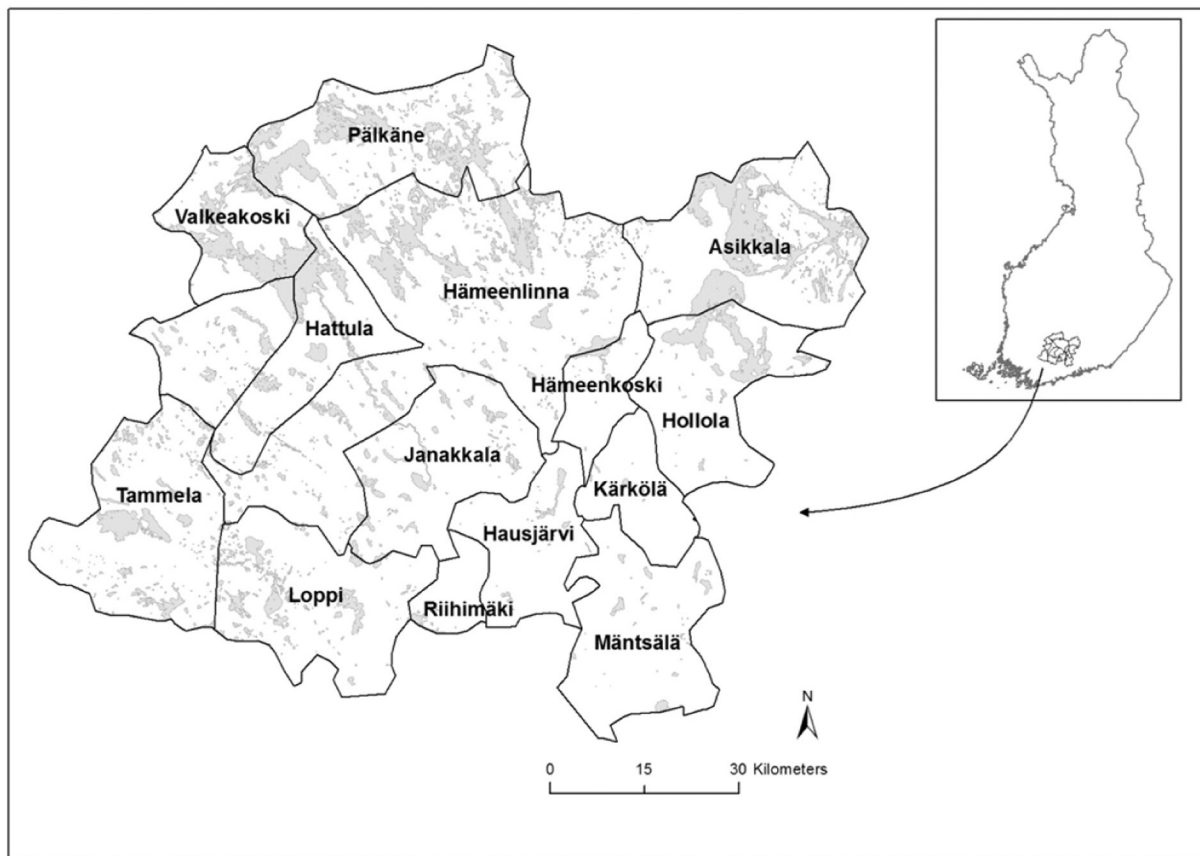


Fig. 1. The study area consisting of 14 municipalities.

2013). The soil area division within municipalities was obtained from Multi-Source National Forest Inventory (Tomppo, 1993) raster maps of 2009. N<sub>2</sub>O emissions from forest fertilisation were calculated based on an estimate of the amount of fertilisers used. The area of fertilised forest in a municipality was estimated based on the total area of fertilised forest in the region.

### 2.2.2. Undrained mires

The carbon budget of undrained mires was calculated based on the net flux estimates by Saarnio et al. (2007). In this study the undrained mires were assumed to be equivalent to pristine mires. The distribution of different regional mire complex types shows that ombrotrophic bogs are the dominant mire types in Southern Finland (Tolonen and Turunen,

1996). Therefore, all the mires in this study were assumed to be ombrotrophic, and emission values of ombrotrophic mires were used.

Emissions from peat extraction sites were calculated using the method used by Finland's national greenhouse gas inventory (Statistics Finland, 2013). To calculate CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O the areas of peat extraction sites were multiplied by emission factors used in the inventory (Statistics Finland, 2013).

### 2.2.3. Croplands

The carbon budget of the croplands includes, as per the national greenhouse gas inventory (Statistics Finland, 2013), a carbon budget of plant biomass and soils and emissions from liming. For the main crops such as grains, root crops and grass, the assumption that the same amount of carbon is sequestered and released annually was

**Table 1**  
Studied municipalities, total areas and areas by land use classes used in the study (km<sup>-2</sup>).

Municipality	Area	Population density people/km <sup>2</sup>	Water	Forest	Cropland	Undrained mires	Peat extraction sites
Asikkala	755	11	192	439	81	7	0.00
Hattula	427	23	70	262	63	14	1.04
Hausjärvi	398	22	9	226	126	9	0.00
Hollola	531	42	69	305	115	8	0.70
Hämeenkoski	195	11	8	126	52	2	0.00
Hämeenlinna	2031	33	246	1392	265	40	0.47
Janakkala	586	29	39	363	130	12	3.34
Kärkölä	259	18	3	150	89	8	0.01
Loppi	655	13	58	470	73	15	0.66
Mäntsälä	596	34	15	368	154	16	0.05
Pälkäne	738	9	178	442	73	11	0.02
Riihimäki	125	233	5	70	25	2	1.79
Tammela	715	9	75	494	88	41	1.61
Valkeakoski	371	57	100	173	59	5	0.02
Study area	8382		1067	5280	1393	190	9.71

made and the effect on the carbon budget is therefore neutral. However, the carbon budget of biomass for woody plants such as apple trees and currant bushes was calculated using the mean of the carbon stock change during past 20 years as a coefficient for biomass C sequestration.

The carbon budget of mineral cropland soils was calculated based on an estimate of the carbon storage in soils and the rate of change in C concentration by Heikkinen et al. (2013). Area unit-based coefficients were formed from this data and were specific to different soil classes, regions and management practices (Viljavuuspalvelu, 2014).

Emission factors for calculating the emissions of organic cropland soils and emissions from liming were obtained from the national greenhouse gas inventory (Statistics Finland, 2013). The shares of different cropland soil types in municipalities were used to divide areas into coarse mineral soils, clay soils and organic soils. Areas of croplands in municipalities divided by crop type and soil type were then multiplied by emission factors. Grasslands are not calculated as a separate land use class; they are included in the cropland category. The area of grassland in the study was 10% of cropland (Statistics Finland, 2013).

#### 2.2.4. Water bodies

The carbon budget calculations for lakes were based on a regionally representative, randomly selected lake data base of Finnish lakes that consists of data from 200 lakes with an area < 100 km<sup>2</sup> (Kortelainen et al., 2006; Juutinen et al., 2009) and all lakes larger than 100 km<sup>2</sup> (37 water bodies) (Rantakari and Kortelainen, 2005). The net gas flux from lakes was calculated as the sum of the net evasion for each lake size class. The size classes considered were lakes smaller than 0.1 km<sup>2</sup>, 0.1–1, 1–10, 10–100, and lakes larger than 100 km<sup>2</sup>. For each size class, the net gas flux was calculated by multiplying the total area of lakes with a net evasion coefficient. The net evasion coefficient refers to the difference between carbon evasion and carbon accumulation to sediments.

The total area of lakes within each size class was used as input data for calculations. The coefficients used for the calculations were based on means of longer time periods and do not take other factors that also have an effect on the carbon gas fluxes into consideration, such as variation in weather conditions, temperature or nutrient levels (Juutinen et al., 2003; Bergström et al., 2007; Juutinen et al., 2009). Nevertheless, the sampling years represent the average annual precipitation patterns in Finland well, and can thus be considered representative for average landscape comparisons.

The values for net CO<sub>2</sub> evasion from lakes per surface area unit were based on a study by Kortelainen et al. (2006). Area-based estimates for CH<sub>4</sub> diffusive flux in the same lake population and similar lake size classes were formed based on the data in Juutinen et al. (2009). CH<sub>4</sub> emission via ebullition was estimated using an equation for CH<sub>4</sub> ebullition per lake (Bastviken et al., 2004) and a sample of lakes in Southern Finland. The data was used to form coefficients for CH<sub>4</sub> ebullition flux per area unit.

CO<sub>2</sub> emission estimates for rivers were based on river areas and the estimated CO<sub>2</sub> fluxes for individual stream orders by Humborg et al. (2010). Rivers were classified by stream order, CO<sub>2</sub> flux and the average width of the rivers in each size class. The total surface area of rivers within each size class was multiplied by the flux value for the respective classes to obtain the total emissions. Further details on the calculation methods used in LUONNIKAS are given in Haaspuro (2013).

#### 2.3. Anthropogenic GHG emissions

Values for anthropogenic GHG emissions on a municipal level were obtained from Finnish Energy Authority and Statistics Finland (2013) databases. The Energy Authority compiles statistics on monitored emissions of combustion installations within the Emission Trading System (EU ETS) (Ellerman and Buchner, 2007). Statistics Finland provides municipal-level emissions data on activities that are not monitored under the EU ETS. To get the total amount of GHG emissions by

municipality, this data was combined. Data for 2010 was used, as regional data for earlier years was not available. Anthropogenic emissions at the national scale in Finland varied mainly due to winter weather conditions from 67 to 80 Mt CO<sub>2</sub>-eq between years 2006 and 2011 and the average value was 73 Mt CO<sub>2</sub>-eq (Statistics Finland, 2013). The value for year 2010 was 75 Mt CO<sub>2</sub>-eq which was slightly higher than the average.

The Energy Authority provides a list of combustion installations and the annual GHG emissions for each installation. As bases of emissions and locations of these installations, we calculated the total EU ETS emissions in each studied municipality. In the study area there were nine installations in four municipalities.

Statistics Finland's municipal-level emissions data is based on Finland's national greenhouse gas inventory (Statistics Finland, 2013). The national-level emissions data is allocated to municipalities based on municipal-level activity data. The methodologies used in the inventory follow the guidelines provided by the Intergovernmental Panel on Climate Change (IPCC) (2013). The calculations follow the principles of production-based emissions calculations, where only the emissions produced inside a municipality's geographical borders on defined sectors are included. Emissions due to the consumption of energy that is produced elsewhere or due to the consumption of goods are not included (SVT, 2013). Our GHG emission calculations include four sectors: energy production, industrial processes, agricultural practices and waste management.

The energy and industry sectors incorporate a variety of emissions. The energy sector covers emissions from fossil fuel combustion and fugitive emissions from fuels. Emissions from fuel combustion include direct and indirect GHG emissions, including point sources, transport and other fuel combustion. GHG emissions from industrial processes include emissions from the chemical industry, metal production, the production of mineral products, and other production such as forest and food industries.

Emissions from the agriculture and waste sectors differ from these. Agricultural GHG emissions cover emissions from enteric fermentation of domestic livestock, emissions from manure management, and the burning of agricultural crop residues. The waste sector covers emissions from solid waste disposal sites including solid wastes and municipal and industrial sludge, municipal and industrial wastewater handling plants, uncollected domestic wastewater, and composting. A detailed description of the methodologies used in the national inventory can be found in the National Inventory Report (Statistics Finland, 2013). The report includes all GHG emissions, whereas the emissions from natural systems reported in this study are based only on CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O.

#### 2.4. Uncertainty analyses

We studied the uncertainty of the LUONNIKAS greenhouse gas budgets by varying the values of the flux rates parameters. The parameter values were randomly sampled (N = 100,000) from uniform or normal distributions. For 11 parameters, representing the carbon flux rates of mineral and organic forest soil, minimum and maximum values for the period 1990 to 2010 were obtained from national greenhouse gas reporting (Statistics Finland, 2013). For the remaining 31 parameters, ranges or standard deviations were not available. For them, values were sampled from normal distributions with means obtained from the literature and standard deviations estimated as 20% of the mean values (see Tables A1–A10 in Supplement information). The standard deviation was chosen large enough to create variation in the results, but small enough that parameter values would not change signs in the analysis.

The results of the uncertainty analysis indicate that the flux rate parameter variation does not determine the probability of one municipality being a larger source or sink than another. The relative errors compared to individual runs introduced in the mean values of the net budgets per municipality were not larger than 4%. The main causes of

the differences between the municipalities are the landscape characteristics (type and area of forest, cropland, water bodies, mires) and the annually varying removal of forest biomass.

### 3. Results

#### 3.1. Land use-related GHG emissions and sinks

In the total area of the 14 studied municipalities, croplands, peat extraction sites, water bodies and undrained mires are all emission sources whereas forests are a large carbon sink, turning the land use-related GHG budget negative, resulting in C sequestration into the ecosystem (Fig. 2).

The total study area of 14 municipalities was a carbon sink when considering land use-related emissions and sinks. The annual sink in the area of municipalities was 650 kt CO<sub>2</sub> eq, which corresponds to 78 t CO<sub>2</sub> eq km<sup>-2</sup> and 2.8 t CO<sub>2</sub> eq per capita. In the forest area of the 14 municipalities, the average carbon sink was 302 t CO<sub>2</sub> eq km<sup>-2</sup> t km<sup>-2</sup> whereas the GHG emissions from cropland, mires and water bodies were 381, 195 and 334 t CO<sub>2</sub> eq km<sup>-2</sup>, respectively.

The municipal-level land use-related GHG budgets varied considerably from about 80 kt of CO<sub>2</sub> eq in Asikkala to about -300 kt in Hämeenlinna. The positive value indicates net emissions of greenhouse gases, and the negative value net sequestration, respectively. For most municipalities the land use-related GHG budgets remained negative, indicating the net carbon sinks, but in Asikkala, Hämeenkoski, Kärkölä and Loppi, the budget was positive (Fig. 3). In these municipalities, the forest C sink was weak or even turned to the emission side due to heavy forest fellings during our study year (2009). However, the forest carbon budget was positive only in Asikkala.

#### 3.2. Anthropogenic GHG emissions

Annual anthropogenic GHG emissions from the whole study area amounted to 2100 kt CO<sub>2</sub> eq, which corresponds to 250 t CO<sub>2</sub> eq km<sup>-2</sup> and 9.2 t CO<sub>2</sub> eq per capita. The municipal level anthropogenic GHG emissions varied considerably from 24 kt CO<sub>2</sub> eq in Hämeenkoski to 504 kt in Hämeenlinna. Riihimäki and Valkeakoski have exceptionally high area-based anthropogenic emissions due to their high industrial activity and small area (Fig. 3). Industrial activity can also be seen in the per capita emissions of Valkeakoski, but the differences between municipalities are not as large per capita as they are per area. This reflects the significance of natural landscapes when estimating the regional carbon budgets.

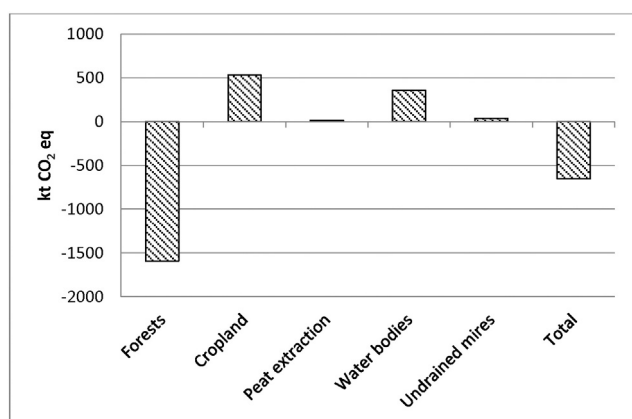


Fig. 2. Land use-related greenhouse gas emissions and sinks of the study area of 14 municipalities.

#### 3.3. Net greenhouse gas budgets

The net land use-related and anthropogenic GHG budget for the study area of 14 municipalities was 1450 kt CO<sub>2</sub> eq, which corresponds to emissions of 172 t CO<sub>2</sub> eq km<sup>-2</sup> and 6.4 t CO<sub>2</sub> eq per capita. The net GHG budgets of the municipalities are shown in Fig. 3. Three municipalities out of 14 were carbon sinks.

The annual anthropogenic GHG emissions from the study area totalled 2100 kt CO<sub>2</sub> eq and the forest sink amounted to 1590 kt CO<sub>2</sub> eq, indicating that 76% of anthropogenic GHG emissions are sequestered into the forests. The three municipalities with significant industrial activity (Hämeenlinna, Riihimäki and Valkeakoski) have the highest levels of anthropogenic emissions. However, Hämeenlinna, with large rural areas, also has a high forest carbon sink. Riihimäki and Janakkala, with their smaller forest areas, have lower carbon sequestration potential.

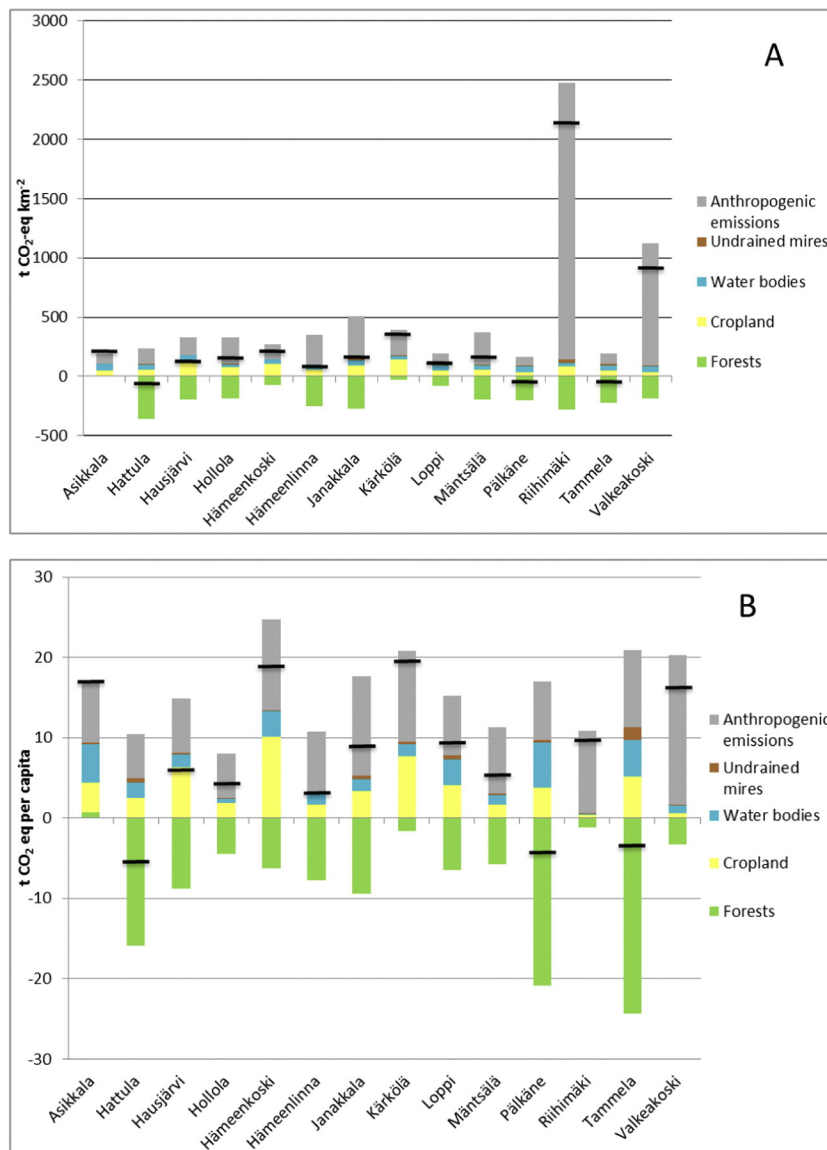
### 4. Discussion

Our results show that even if the cropland, mires and water bodies have higher area-based emissions than area-based forest sinks, the larger area of forest can compensate for these emissions in most of the municipalities. However, the amount of annual cuttings is significant for a municipality's one-year carbon budget. Even if the forest area of a municipality is large and the annual tree growth contributes to considerable C sequestration, substantial cuttings can turn the land use-related carbon budget positive, i.e. the forests, cropland and natural areas of a municipality are a carbon source. Asikkala, which has the fifth-largest forest area of all the municipalities in this study, is the only municipality with a positive carbon budget for forests. In addition, Loppi, which has the third-largest forest area, has a positive total carbon budget of land use-related carbon.

The municipalities' emissions from different natural sources are related to the area of the different environments. Urban areas predominantly represent only a small proportion of the surface area of the municipalities, and large overall areas include relatively large areas used for farming or forestry, or natural environments without any significant human impact. The population density is rather low in all the studied municipalities. Consequently, the surface area covered by forests is most important in these communities when the aim is to reduce GHG emissions and achieve carbon neutrality.

Because the GHG emissions from the lakes, rivers and undrained mires are directly related to the area of these ecosystems in municipalities, their share of the total emissions varied. The share of emissions from undrained mires is rather small in all municipalities' budgets, which can be explained by the relatively small areas of undrained mires in general (draining of peatlands for forestry has been intensive in Finland) rather than low area-based emissions. In some municipalities that have a small area covered by lakes, the role of rivers become more significant in the carbon budget, e.g. in Kärkölä, where almost 90% of the emissions from water bodies comes from rivers, with the total freshwater contribution representing 20% of the total budget. This is in agreement with the recent boreal estimate by Weyhenmeyer et al. (2012), which emphasized significant contribution from river evasion. Global estimates by Raymond et al. (2013) identified hotspots in stream and river evasion, with about 70% of the flux occurring over just 20% of the land surface. Lauerwald et al. (2015) ended up with lower CO<sub>2</sub> evasion estimates for tropical rivers, but highlighted population density as a controlling factor of CO<sub>2</sub> evasion from global river network.

The calculations include uncertainties and are based on average values and leave no room for site-specific variation. Inter-annual and temporal variation in emissions is large due to weather conditions. Precipitation was shown to be a key driver of the variability in aquatic annual C balance in boreal landscapes (Einola et al., 2011) and also explained the global freshwater hotspots (Raymond et al., 2013). In a forest's annual budget, the fellings have a substantial role. Clear cut



**Fig. 3.** Greenhouse gas budgets for municipalities. Net-GHG budgeted is marked with the horizontal line for each municipality. Per area (A) and per capita (B).

felling, which is a normal forest management practice in the study area, removes all the stem wood biomass from the forest and reduces the carbon sequestration significantly. As the calculations are fixed to a specific year (2009), land use changes were not considered.

The results obtained with the LUONNIKAS calculation tool are useful as an initial estimate of the landscape' role in municipal-level carbon budgets, and allow for a comparison of anthropogenic and land use-related emissions. Our study is a first attempt to connect ecosystems' carbon budgets and anthropogenic emissions on a municipal level over such a large area. The LUONNIKAS calculation tool allows for municipal-level calculations of land use-related carbon gases using easily available data, and these calculations can be performed systematically with the same methods in any municipality in Finland, thus also enabling comparable results between municipalities.

Since boreal forests are a significant carbon sink and the efficiency of this sink is heavily impacted by forest management practices, forest management policy is a key contributing factor in mitigating landscape GHG emissions. Further, eutrophic lakes have been shown to evade more CO<sub>2</sub> and CH<sub>4</sub> than oligotrophic lakes (Kortelainen et al., 2006; Juutinen et al., 2009), which underlines the importance of effective waste water treatment and land use policy in the attempt to reduce the load of nutrients and thus to increase C sequestration and reduce

GHG emissions from boreal landscapes. Similarly, emissions from peatlands and croplands can also be influenced by management decisions. Municipalities are thus in a key position to make decisions that affect local C emissions and sinks and are therefore important for GHG mitigation.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.scitotenv.2016.03.040>.

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