The contribution of black carbon to the carbon storage in a permafrost ecosystem

Georg Guggenberger[1,*], Andrej Rodionov[1], Matthias Grabe[1], Oleg A. Kasansky[2],
Olga Shibistova[3], and Heiner Flessa[4]

[1] *Institute of Soil Science and Plant Nutrition, Martin Luther University Halle-Wittenberg, 06099 Halle, Germany*
[2] *Field Station Igarka of the Permafrost Institute Yakutsk, SB-RAS, Microrayon 2a, Igarka, Russian Federation*
[3] *V.N. Sukachev Institute of Forest, Forest Monitoring Laboratory, SB-RAS, Akademgorodok, 660036 Krasnoyarsk, Russian Federation*
[4] *Institute of Soil Science and Forest Nutrition, University of Göttingen, 37077 Göttingen, Germany*

[*] *corresponding author: georg.guggenberger@landw.uni-halle.de*

1. INTRODUCTION

Vegetation fires frequently convert organic matter to black carbon (BC). This term describes a continuum of aromatic carbon with highly condensed structures from partly charred plant material through charcoal to soot with no general agreement on clear-cut boundaries (Schmidt et al., 2001). Due to its condensed structure, BC is believed to be quite refractory and may represent a significant sink in the global carbon cycle (Kuhlbusch, 1998). There is a good number reports showing that BC accumulates in soils of some ecosystems where either fires are frequent (Skjemstad et al., 1996; Schmidt et al., 1999) or where charred residues were added to the soil (Glaser et al., 2001). Estimated turnover rates were in the range of centuries to millennia. However, there are other data, proofing that BC can be decomposed by microbial action (Rethemeier et al., 2005) and photooxidation (Shneour, 1966) which produces highly aromatic acids that are extractable in aqueous solutions (Haumaier and Zech, 1995; Skjemstad et al., 1996; Möller et al., 2000). Hence, BC can get lost from soil by microbial respiration and by dissolution and leaching from soil in addition to soil erosion (Schmidt and Noack, 2000; Mannino and Harvey, 2004). Bird et al. (1999) showed for a well-aerated savanna soil that natural degradation of BC takes place on decadal time scales which suggests that most of the BC cannot be considered to represent a slow-cycling reservoir.

In boreal regions fire is the most important disturbance due to pronounced summer droughts. According to Wirth et al. (2002), about 35% of the net primary production of Siberian Scots pine forests located on the eastern border of the West Siberian Plain is lost by fires. Czimczik et al. (2003) reported that about 0.7 – 8% of the carbon is converted to BC, which can potentially accumulate in soil. But the BC stocks in the sandy, well-aerated soils below the Siberian Scots pine are surprisingly small. Czimczik et al. (2005) showed that BC in these soils ranged between 0.1 and 72 g m^{-2}. Hence, black carbon did not significantly contribute to the storage of organic matter in these forests, most likely because it is completely oxidised to CO_2 during intensive fires (Czimczik et al., 2005).

Yet, there are no investigations about the fate of BC in permafrost ecosystems. Due to the shorter vegetation period with smaller evapotranspiration, summer droughts are not as intensive in the northern taiga and the forest tundra as in the central taiga. Hence, fire frequency is smaller. However, fire disturbance may be quite important in permafrost ecosystems as well, since fire may initiate permafrost melting and formation of thermokarst (Yoshikawa et al.,

Symptom of Environmental Change in Siberian Permafrost Region,
Eds. Hatano R and Guggenberger G, p 75-84, Hokkaido University Press, Sapporo, 2006

2002). Here, we report about a comparative analysis of total soil organic carbon (OC) and BC storage in a small catchment at the forest tundra ecotone at the lower Yenissej. The goal of our research was to analyze the contribution of BC to OC in different landscape units and in particular to assess the relation of BC stocks in soil to the permafrost regime.

2. MATERIALS AND METHODS

2.1 Study area

The study was carried out in the 0.44 km^2 sized Grawijka Creek catchment located approximately 10 km north of the town of Igarka (Russian Federation) at 67^{o}29.90’ latitude and 86^{o}25.26’ longitude on the eastern shore of the Yenisei river. The region is situated in the transition zone from the moderately continental western Siberian climate to the strongly continental eastern Siberian climate. In nearby Igarka the average annual temperature is -7.4 ^{o}C and the mean annual precipitation is about 510 mm with about half of it falling during the summer months July, August, and September (Lieth et al., 1999). Snow cover usually extends from end October to mid May.

The study site is located in the forest tundra ecotone. The vegetation is a mixed forest dominated by *Larix sibirica*, *Picea obovata*, *Betula pubescens*, and *Pinus sibirica*. Stands are relatively dense in areas with lacking or low permafrost, whereas the areas with high permafrost are characterised by open stands. Ground vegetation is generally characterized by *Vaccinium myrtillus*, *Vaccinium uliginosum*, *Rubus chamaemorus* and various grasses on well drained soils and is dominated by *Sphagnum* mosses at wet sites.

Parent materials in the catchment area are Quaternary sediments derived primarily from the Karginskaya interstage (59 - 24 kyrs BP) which is of glaciofluvial to glaciolimnic genesis and characterized by silty deposits (Sachs, 1948). According to the Geocryological Map of the USSR, the Grawijka Creek catchment is located within the continuous permafrost belt (Ershov, 1996). Own observations suggest that the topographic position and associated soil drainage and ground vegatation are important local factors determining distribution and depth of permafrost in the catchment (Rodionov et al., submitted). In fact, well-drained south southwest (SSW) facing slopes show thick active layer or lacking permafrost, while north northeast (NNE) exposed slopes have wet and partly peaty soils with a higher permafrost table. The plateau areas are covered by bogs which are having a thin active layer. Within this bog area, permafrost degradation by thermokarst erosion with the formation of thermokarst ponds and gullies could be frequently observed. Based on these observations, Rodionov et al. (submitted) distinguished four landscape units: SSW facing slopes, NNE exposed slopes, intact bogs, and degraded bogs.

2.2 Soil sampling and analysis

From June to July 2003 a 50 x 50 m grid mapping of the catchment was carried out. Based on these grids a total number of 168 points were defined in the catchment. At these points the active layer depth was measured, and soil samples were taken by volume from the organic layer/moss layer, and the mineral soil 0-20 cm, 20-50 cm, and 50-90 cm depth increments using a soil corer. Decomposed organic horizons of the Histosols were collected likewise.

Organic layer/moss layer and mineral soil samples were air-dried immediately after sampling. Prior to analyses, mineral samples were passed through a 2-mm sieve and organic samples were ground using a ball mill. Bulk density was determined by weighing of the air-dried volume samples. Total carbon was determined by dry combustion with an Elementar Vario Maxi CHNS analyser (Elementar Analysensysteme GmbH, Hanau, Germany). Most soil samples were free of carbonates, however, some contained traces of carbonates (< 1 mg

$CaCO_3$-C g^{-1} soil), which were subtracted from the total carbon in order to obtain OC. All analyses were performed in duplicate; the coefficient of variation was <1.7%.

Black carbon was analysed using benzenepolycarboxylic acids (BPCA) as molecular markers (Glaser et al., 1998 in the modified version of Brodowski et al., 2005). Briefly, this methods includes a digestion with 4 M trifluoroacetic acid (TFA) to remove most polyvalent cations from the mineral soil samples. The benzenepolycarboxylic acids (hemimellitic, trimellitic, trimesic, pyromellitic, prehnitic, mellophanic, benzenepentacarboxylic and mellitic acid) were released from the condensed original BC structure by oxidation with HNO_3. The BPCAs were purified by a cation exchange resin and analyzed as trimethylsilyl (TMS) derivatives by capillary gas chromatography (GC). The GC analysis was performed on a Shimadzu GC - 2010 gas chromatograph (Shimadzu, Jena, Germany) equipped with a flame-ionization detector (FID) and a SPB-5 capillary column (30 m x 0.25 mm i.d., 0.25 µm film thickness, Supelco, Nr. 24034). The sum of the yields of prehnitic, mellophanic, benzenepentacarboxylic and mellitic acid is a relative measure of the BC in the soil (Glaser et al., 1998). All analyses were performed in duplicate; the coefficient of variation averaged 10%. The real amount of matter is difficult to assess because BC exists as a continuum of thermally altered material, whereas analytical methods rely on operational definitions with clear-cut boundaries (Schmidt et al., 2001).

2.3 GIS processing and statistical analysis

Geographic Information Systems (GIS) were used for the creation of maps that show the spatial variation of data points. We employed the GIS-software ArcView GIS Versions 3.2 including the extension Spatial Analyst (ESRI, 1999). The map layout was done using the GIS-software ArcGIS 8.1 (ESRI, 2001). The two-dimensional OC map were created by interpolating point data from the 168 sampling points to the area. Because BC analysis is a very laborious and time-consuming process, in case of the BC the map was based on 48 sampling points only,. Interpolation was done by creating a grid with a cell size of 0.5 x 0.5 m using the interpolation method spline in the Spatial Analyst.

In order to relate OC and BC storage to environmental parameters, a canonical correspondence analysis was carried out (Ter Braak, 1986). In this statistical analysis ordination axis are chosen in the light of known environmental variables by imposing the extra restriction that the axes be linear combinations of environmental variables. For each axis an eigenvalue is given that will be reached at maximum distance of the variables for the axis. Hence, it is a measure for the impact of the axis. In the canonical correspondence analysis $cosine^2$ (quality or squared correlations with each dimension) contains the quality for each point, by dimension. The sum of the values across the dimensions defines the total quality value. This value may also be interpreted as the correlation of the respective point with the respective dimension. Since this method is an exploratory technique, no statistical significance tests are customarily applied to the results (Greenacre 1984).

3. RESULTS AND DISCUSSION

3.1 Total organic carbon stocks

Soil organic carbon stocks in the Grawijka Creek catchment to a depth of 90 cm (plus organic layer/moss layer) varied strongly between 5 and 95 kg C m^{-2} (Fig. 1). Largest OC stocks were observed in the intact bogs (average 52 kg C m^{-2}), followed by the NNE exposed soils (28 kg C m^{-2}), the degraded bogs (26 kg C m^{-2}), and the SSW faced soils (22 kg C m^{-2}). The size of OC stocks in the Grawijka Creek catchment fits well to other studies on OC stocks in permafrost soils. Organic carbon stocks in the upper meter of permafrost soils of Alaska ranged from 17 to 130 kg m^{-2} (Ping et al., 1997). There, largest SOC storage of up to 130 kg m^{-2} was found in bogs, arctic tundra soils had stored 31 to 60 kg C m^{-2}, and the SOC content of forest pedons ranged from 17 to 79 kg m^{-2}. In another study in Alaska, Michaelson et al. (1996) reported OC stocks from 16 kg m^{-3} for a northern foothills ridge top tundra site to 94 kg SOC m^{-3} for a Barrow coastal plain tundra. Hence, it appears that the spatial variability of SOC stocks in the 0.44 km^{2} Grawijka Creek catchment is as large as in the much more large-scale studies in Alaska, indicating that local factors have an impact on SOC storage at a same order of magnitude than regional ones.

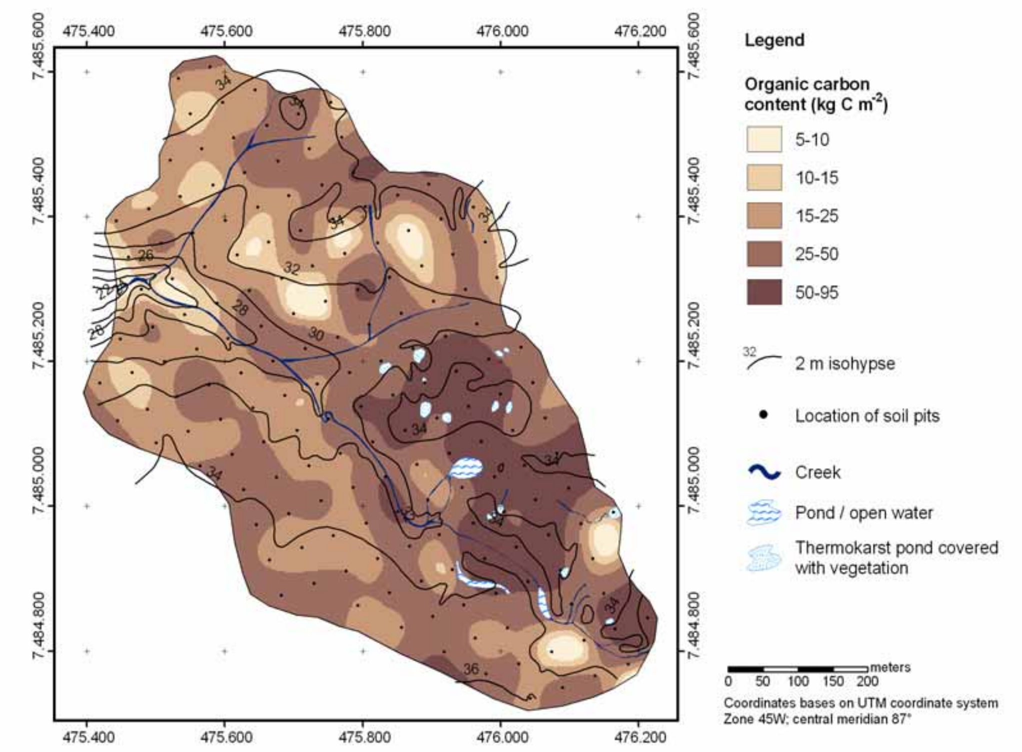

Fig. 1. Total organic carbon stocks in the Grawijka Creek catchment in the organic layer/moss layer and the 0-90 cm mineral soil/organic soil horizons in kg m^{-2}.

The canonical correspondence analysis suggests closest relation of the OC stocks to the active layer thickness among all environmental parameters tested (not shown). Organic carbon stocks of 0-10 kg m^{-2} show a canonical correspondence coefficient cosine^{2} with the active layer thickness of –0.94, while for the OC stocks 50-95 kg m^{-2} with the active layer thickness it is 0.79. This indicates that high OC stocks concur with thin active layers while low OC stocks are found in soils with thick active layers. Rodionov et al. (submitted) further showed that soils with shallow active layers store more OC in all depth increments than soils with deep active layer or lacking permafrost. Within the permafrost, organic matter is preserved by freezing. Further, soils with thin active layers exhibit a limited drainage, in particular at plains and gentle slopes, leading to the formation of soils with gleyic properties (Guggenberger et al., 2001). The resulting anaerobiosis in the active layer retards organic matter decomposition (Hobbie et al., 2000).

Differences in the OC stocks between the NNE and SSW faced mineral soils are not very pronounced. Obviously, the more favourable conditions for organic matter mineralisation in the

SSW exposed soils caused by higher soil temperature and drier soil conditions is possibly counterbalanced by the higher net primary production due to the more favorable soil chemical and physical conditions. In contrast, differences between the OC stocks between the soils in intact and degraded bog areas are by a factor of two. This implies that thermokarst-induced permafrost degradation in the raised bogs due to climatic or other disturbances may be a crucial process in the OC storage in these landscape units. One possible disturbance being responsible for thermokarst formation is fire. In the following we will test whether BC, which can be considered as a proxy of fire history (Bird and Cali, 1998), indicates forest fire as a possible reason of permafrost degradation.

3.2 Contribution of black carbon to total organic carbon

According to Fig. 2, black carbon as identified by the BPCA method is ubiquitous in the soils of the catchment, it can be found at any soil depth, and contributed to about 0.3 – 3% to the total soil organic carbon. Soil organic carbon in the intact bogs had higher proportions of BC than soils of the other landscape units.

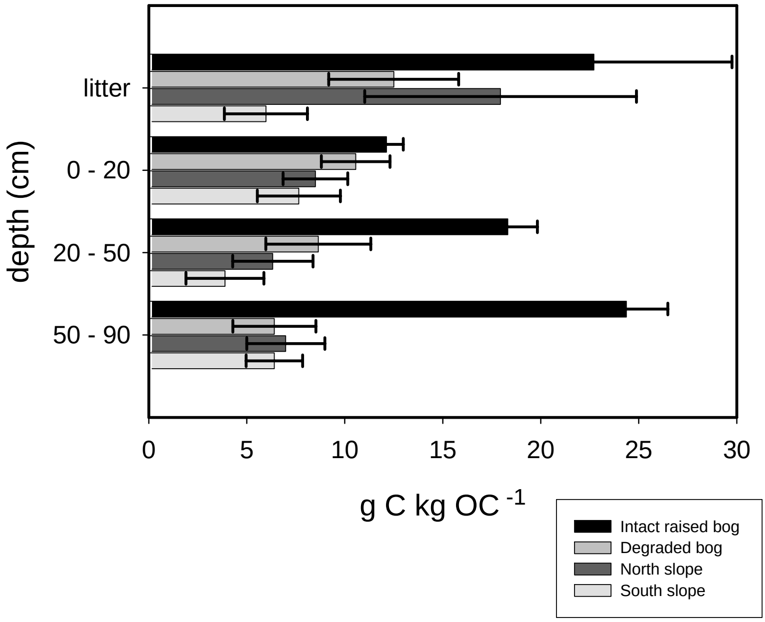

Fig. 2. Contribution of black carbon to the total organic carbon (g C kg OC^{-1}) in soils of the different landscape units.

Here, it has to be noted that the BPCA method, as any method to determine BC, only addresses a certain window of the BC continuum (Hammes et al., submitted). Brodowski et al. (2005) showed that highly condensed graphitized BC may have survived oxidation and conversion into BPCA. On the other hand, wood and grass char may have a low degree of aromatic condensation and will, therefore, not produce penta- or hexacarboxylated benzoic acids (Brodowski et al., 2005). Further, the conversion of BC to BPCAs is incomplete in general, since the carboxylic groups are formed from former aromatic neighbours. We also omitted to use a correction factor as was obtained from a set of BP standard materials by Glaser (1999), since the degree of aromatic condensation of BC in the soil is not known. Hence, the

BC concentrations reported here are conservative estimates and the “true” proportion of BC on OC is expected to be higher than indicated by the BPCA method. This complicates also a comparison with the BC concentrations in other soils, which is in fact only valid if the same method was used. Glaser and Amelung (2003) reported that BC in native grassland soils in North America, where fire frequency is much higher as it is in the Siberian forest tundra, contributed between 4 and 18% to OC. For soils located at the eastern edge of the West Siberian Plain, Czimczik et al. (2005) showed a contribution of <3.5% BC to OC in the litter layer below Siberian Scots pine, which is similar to values observed in the litter layer of the soils further north in the Grawijka Creek catchment.

In the top mineral soil, however, BC accounted only to 0.02% of OC in the well-aerated sandy soils investigated by Czimczik et al. (2005). In contrast, the soils in the Grawijka Creek catchment also showed significant concentrations at greater soil depth (Fig. 2). This is in particular true for the 20-50 and 50-90 cm depth increments in the intact bogs, which are usually permanently frozen. The layered stratigraphy of the peat suggests that not cryoturbation was the reason for the translocation of BC to greater soil depths. It rather appears that with the continuing growth of the peat after a fire event, burnt residues got buried below the moss layer and with time got sequestered in the upwards growing permafrost. The situation might be different for the mineral soils of the catchment. Also there, albeit at a lower extent, significant a BC concentration can be found in deeper soil layers. A possible reason may be biotic or abiotic oxidative degradation of BC rendering the molecules more water soluble. So Möller et al. (1999) showed occurrence of BP-derived free mellitic acid in an aqueous extract of a subsoil of a tropical forest ecosystem in Thailand. In the BPCA method the ratio of benzenepentacarboxylic acid to mellitic acid (b5ca/b6ca) can be used as an indicator of the degree of aromaticity and/or degree of decomposition of BC (Glaser, 1999). Increasing ratios suggest that an increasing proportion of the neighbouring carbon is not aromatic. The values reported in Fig. 3 are about twice as high as those found in an anthropogenic soil in Brasil

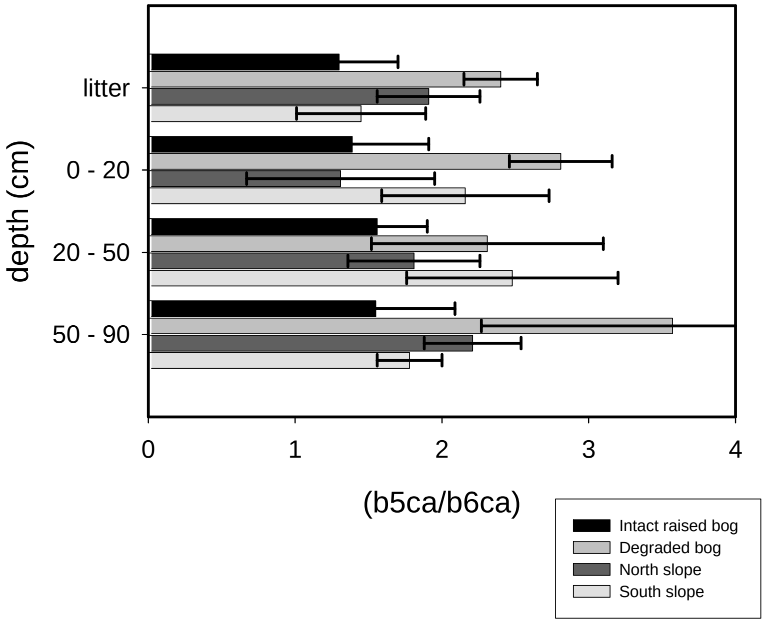

Fig. 3. Ratios of the benzenepentacarboxylic acid (b5ca) to mellitic acid (b6ca) in soils of the different landscape units.

(Glaser, 1999). Hence, BC molecules may be considered to contain functional groups rendering them mobile in the soil profile. Striking is the difference in the (b5ca/b6ca) ratio in the soils of the intact and degraded bogs, which is twice as high for the latter. In combination with the smaller contribution of BC to OC (Fig. 2), this suggests that degradation of the bogs by permafrost thawing may lead to decomposition and mobilisation of BC.

3.3 Black carbon stocks

Black carbon stocks in the Grawijka Creek catchment to a depth of 90 cm (plus organic layer or moss layer, respectively) varied strongly between about 50 and 4.000 g C m^{-2} (Fig. 4).

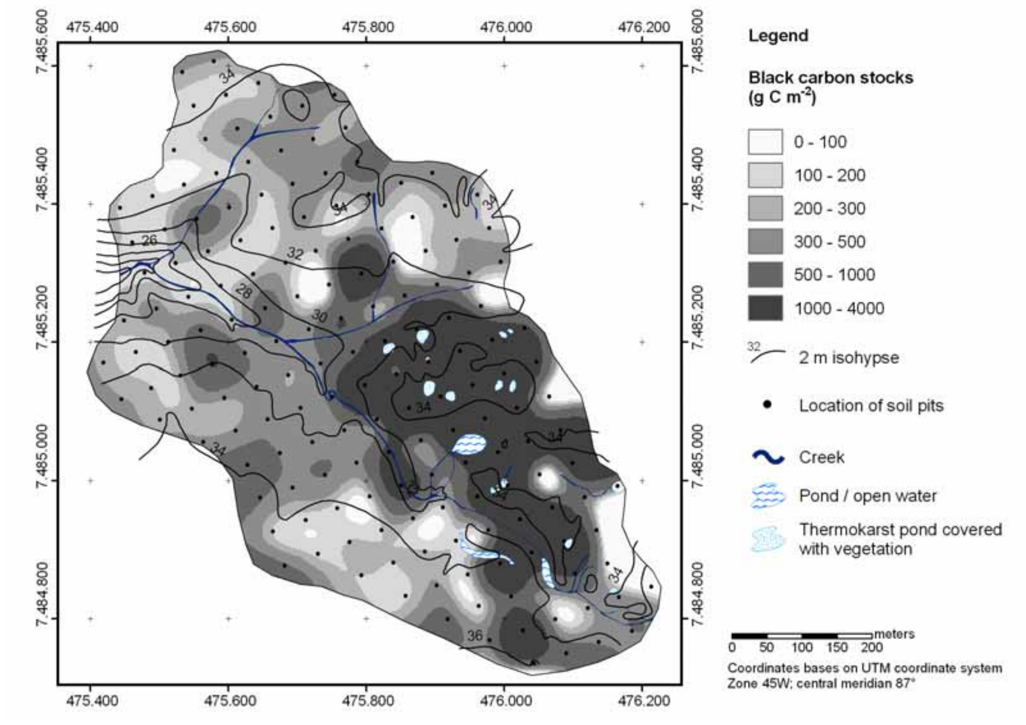

Fig. 4. Black carbon stocks in the Grawijka Creek catchment in the organic layer/moss layer and the 0-90 cm mineral soil/organic soil horizons in g m^{-2}.

Largest stocks were observed for the intact bogs with average values of about 1.500 g C m^{-2}. Sixty eight percent of the BC is located within the permafrost, where it is possibly stabilised against decomposition and leaching. In the degraded bogs, having no permafrost, average BC stocks were only about 390 g C m^{-2}. The smaller storage of BC in the degraded bogs as compared to the intact bogs is due to a smaller storage of total organic carbon (Fig. 1) and due to the smaller proportion of BC to OC (Fig. 2). Mineral soils stored generally less BC than the bogs with 400 g C m^{-2} in the NNE facing soils and 230 g C m^{-2} in the SSW exposed soils. Also for the mineral soils it appears that those with more abundant permafrost and a thinner active layer store more BC. To verify this indication, we carried out a canonical correspondence analysis.

In Fig. 5 the BC stocks of the soils within the catchment are related to the environmental variables landscape unit, active layer thickness, and OC stocks by the aid of a canonical correspondence analysis. Highest BC stocks (BC 5) are associated with intact raised bogs, active layer thickness <20 cm, and highest OC stocks (OC 5). The second group contains intermediate BC stocks (BC 3, BC 4) that can be typically found in NNE oriented slopes of intermediate active layer thickness. This group is further characterised by intermediate OC stocks. Smallest BC stocks are occurring in one group including SSW aspects and the degraded raised bog. This group is also characterised by deep or lacking permafrost and smallest OC stocks. Also the canonical correspondence coefficient cosine^2 reveals that the largest BC stocks are closely related to the integrated bogs (0.82), to the largest OC stocks (0.78), and to the least active layer thickness (0.45) (not shown).

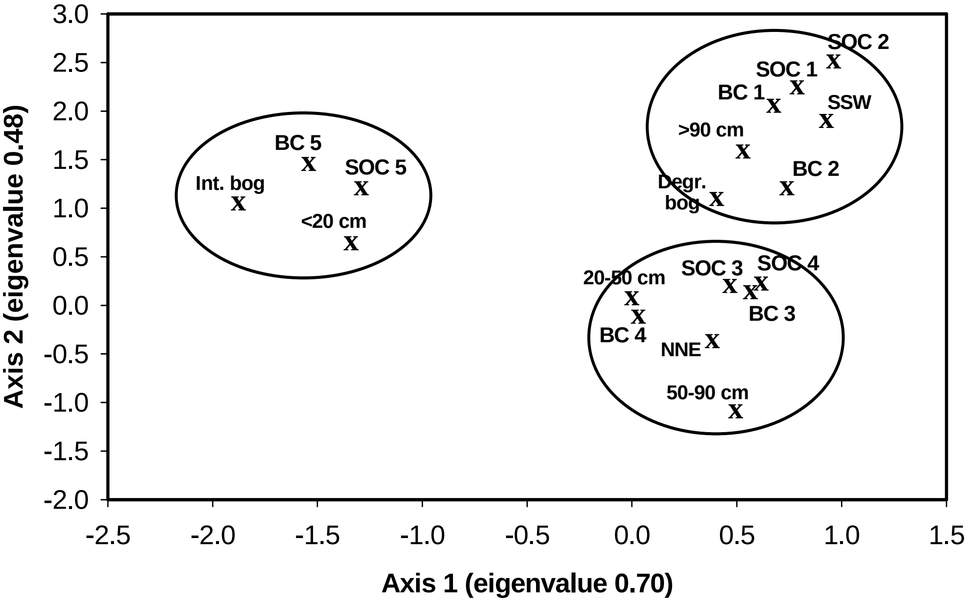

Fig. 5. Canonical correspondence analysis ordination for black carbon stocks (g m^{-2}) with landscape unit, active layer thickness and soil organic carbon stocks as variables. Annotations: black carbon stocks: BC 1, 0-100 g C m^{-2}; BC 2, 100-200 g C m^{-2}; BC 3, 200-500 g C m^{-2}; BC 4, 500-1.000 g C m^{-2}; BC 5, 1.000-4.000 g C m^{-2}; landscape units: Int. bog, intact bog; Degr. bog, degraded bog; NNE, north northeast slope; SSW, south southwest slope; active layer thickness: <20 cm, 20-50 cm, 50-90 cm, >90 cm; soil organic carbon stocks: SOC 1, 5-10 kg C m^{-2}; SOC 2, 10-15 kg C m^{-2}; SOC 3, 15-25 kg C m^{-2}; SOC 4, 25-50 kg C m^{-2}; SOC 5, 50-95 kg C m^{-2}.

The results of this study thus provides evidence that the BC stocks are strongly related to the OC stocks in general and to the environmental variables that control the OC stocks. A similar result was obtained by Glaser and Amelung (2003) for BC in North American native grassland soils. They concluded that the BC storage in soils is controlled by higher BC production at sites with high plant biomass and by shorter BC residence time in soils showing favourable conditions for OC decomposition. In the intact bogs of the Grawijka Creek catchment most of the BC is stored in the permafrost, suggesting that this preservation mechanism is not only important for OC but also for BC. And the fact that most BC can be found in the soils with high permafrost table further shows that fire does not necessarily lead to permafrost degradation. Obviously, past vegetation/peat fires in the Grawijka Creek catchment were not very intense and burnt only the upper part of the moss and peat layers. After the fire event, mosses continued to grow and buried the burnt residues which got sequestered with time in the upwards growing permafrost. For the mineral soils, a different permafrost regime have not such a pronounced influence on the BC storage. But also for the mineral soils it appears that BC is rather sequestered within the permafrost than being an indicator of fire-induced permafrost degradation.

The data also show that if permafrost degradation is taking place by some kind of disturbance, in particular the bogs will loose BC to a large extent. It is proportionally even more pronounced than for OC in general. Hence, BC in permafrost soils is highly susceptible to permafrost thawing and cannot be considered as a refractory carbon species in such ecosystems. But the pathways of these losses are not clear yet. It is unlikely that complete thermic oxidation of BC during intense fire events is the reason, as was shown by Czimczik et al. (2005) for well aerated soils further south, because most of the BC investigated in the present study is stored at larger soil depths. Losses by soil erosion (Schmidt and Noack, 2000) may explain mass losses of BC in thermokarst erosion gullies but not the preferential losses of BC over OC in degraded bogs themselves. At the moment, we don’t have any idea about the relevance of decomposition/humification of BC and leaching of BC in these soils. As these losses are concurring with decreasing (b5ca/b6ca) values, disintegration and oxidation of the BC molecules and further leaching from the soil into the hydrosphere might be a possible pathway (Möller et al., 1999; Mannino and Harvey, 2004). This possibility will be investigated in ongoing research.

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