

1 **Carbon and greenhouse gas balances in an age-sequence**
2 **of temperate pine plantations**

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1 **Abstract**

2 This study investigated differences in the magnitude and partitioning of the carbon (C) and
3 greenhouse gas (GHG) balances in an age-sequence of four white pine (*Pinus strobus* L.)
4 afforestation stands (7, 20, 35 and 70 years old as of 2009) in southern Ontario, Canada. The
5 4-year (2004-2008) mean annual carbon dioxide (CO₂) exchanges, based on biometric and
6 eddy covariance data, were combined with the 2-year means of static chamber measurements
7 of methane (CH₄) and nitrous oxide (N₂O) fluxes (2006-2007) and dissolved organic carbon
8 (DOC) export below 1 m soil depth (2004-2005). The total ecosystem C pool increased with
9 age from 46 to 197 t C ha⁻¹ across the four stands. Rates of organic matter cycling (i.e. litter-
10 fall and decomposition) were similar among the three older stands. In contrast, considerable
11 differences related to stand age and site quality were observed in the magnitude and
12 partitioning of individual CO₂ fluxes showing a peak in production and respiration rates in the
13 middle-age (20-year-old) stand growing on fertile post-agricultural soil. The DOC export
14 accounted for 10% of net ecosystem production (NEP) at the 7-year-old stand but <2% at the
15 three older stands. The GHG balance from the combined exchanges of CO₂, CH₄ and N₂O
16 fluxes was 2.6, 21.6, 13.5 and 4.8 t CO₂ eq ha⁻¹ y⁻¹ for the 7, 20, 35 and 70-year-old stands,
17 respectively. The maximum annual contribution from the combined exchanges of CH₄ and
18 N₂O to the GHG balance was 13% and 8% in the 7 and 70-year-old stands, respectively, but
19 <1% in the two highly productive middle-age (20 and 35-year-old) stands. Averaged over the
20 entire age-sequence, the CO₂ exchange was the main driver of the GHG balance in these
21 forests. The cumulative CO₂ sequestration over the 70 years was estimated at 129 t C and 297
22 t C ha⁻¹ y⁻¹ for stands growing on low and high productive sites, respectively. This study
23 highlights the importance of accounting for age and site quality effects on forest C and GHG
24 balances. It further demonstrates a large potential for net C sequestration and climate benefits
25 gained through afforestation of marginal agricultural and fallow lands in temperate regions.

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1 **1 Introduction**

2 The global temperature increase over the past century has been attributed to increasing
3 concentrations of atmospheric greenhouse gases (GHGs) such as carbon dioxide (CO₂),
4 methane (CH₄), and nitrous oxide (N₂O), among others, coupled with human activities (e.g.
5 fossil fuel burning, deforestation) and natural processes in terrestrial ecosystems (Houghton et
6 al., 1998; IPCC, 2013; Schimel, 1995; Schulze et al., 2009). Much effort has since been put
7 into mitigating the atmospheric GHG concentrations through sequestration and/or their
8 emission reduction strategies through land use management activities (Montzka et al., 2011;
9 Nabuurs et al., 2007; Smith et al., 2008).

10

11 Among land use types, forest ecosystems have the strongest carbon (C) sink potential and
12 provide stocks of 100 to 160 Gt C in biomass and soil in temperate regions alone (Dixon et
13 al., 1994; Goodale et al., 2002; Pan et al., 2011; Reich, 2011). Moreover, much of the current
14 CO₂ sink strength of the northern hemisphere has been attributed to re-growth and
15 afforestation of former agricultural land in the US and Canada (Pan et al., 2011; Reich, 2011).
16 Therefore, further afforestation of former agricultural lands of low productivity has been
17 proposed to help offset the increasing concentration of atmospheric CO₂ (Bárcena et al., 2014;
18 Brown, 2002; Goodale et al., 2002; Nabuurs et al., 2007; Niu and Duiker, 2006).

19

20 Afforestation of former agricultural lands and associated changes in land management
21 practices, however, does not only result in accumulation of C into biomass but may also alter
22 soil and micro-climatic conditions. Tree root development, litter-fall, canopy shading and
23 cessation of N-fertilizer application, for instance, may trigger changes in physical,
24 biogeochemical and hydrological properties of the soil which may affect the net exchange of
25 CH₄ and N₂O (Ball et al., 2007; Christiansen and Gundersen, 2011; Christiansen et al., 2012;
26 Gundersen et al., 2012; Peichl et al., 2010b; Priemé et al., 1997; Smith et al., 2003; Ullah and
27 Moore, 2011), as well as the cycling of dissolved organic carbon (DOC) (Camino-Serrano et
28 al., 2014; Gielen et al., 2011; Peichl et al., 2007; Rosenqvist et al., 2010). Such alterations
29 might modify the net ecosystem production (NEP) and the GHG balance (Luyssaert et al.,
30 2010, 2012; Schulze et al., 2009, 2010). Moreover, since CH₄ and N₂O have a 34 and 298
31 times greater global warming potential (GWP) relative to CO₂ on molar basis over 100 years
32 when including carbon-climate feedbacks (IPCC, 2013), afforestation effects on the exchange

1 of these GHGs might be even more pronounced with respect to the net radiative forcing
2 (Inatomi et al., 2010; Montzka et al., 2011; Schulze et al., 2009).

3

4 The impacts of afforestation on ecosystem C pools and GHG fluxes may also change with
5 stand age over time, including short-term (<10 years; Bjarnadottir et al., 2009; Don et al.,
6 2009; Zona et al., 2013), intermediate (~10 to 50 years; Ball et al., 2007; Christiansen and
7 Gundersen, 2011) and long-term (>50 years; Coursolle et al., 2012; Hiltbrunner et al., 2012;
8 Priemé et al., 1997) effects. In addition, site quality (which includes all environmental factors
9 influencing tree growth and thus biomass production and decomposition) might exert a strong
10 control on the forest CO₂ exchange and C flux partitioning (Fernández-Martínez et al., 2014;
11 McLeod and Running, 1988; Peichl et al., 2010a; Vanninen et al., 1996; Vicca et al., 2012).
12 Changes in the magnitude of the CO₂ exchange subsequently has implications for the relative
13 contribution of the individual C and GHG fluxes to the total C and GHG budgets following
14 afforestation. Thus, knowledge on the changes over the entire life span of the afforested stand
15 and on the initial site conditions and quality are required to understand the overall
16 implications of such land use practices for the C and GHG balances at ecosystem and regional
17 scales.

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19 To date, few attempts have been made to estimate the full forest C and GHG balance by
20 including measurements of all relevant fluxes of CO₂, CH₄, N₂O, and DOC. Ball et al. (2007)
21 estimated that the combined contribution of CH₄ and N₂O to the total GHG balance was 6-7%
22 in maturing (20-30 years old) coniferous forests. In a young poplar plantation, the
23 contribution of CH₄ and N₂O was reported to account for 50% of the GHG balance (Zona et
24 al., 2013). Using a process-based model, Inatomi et al. (2010) estimated the GHG balance of a
25 50-year-old temperate broadleaved forest at 10 t CO₂ ha⁻¹ y⁻¹ by including all three GHGs
26 (CO₂, CH₄, N₂O), with a small (<2%) contribution from CH₄ and N₂O fluxes. On a larger
27 regional scale, Tian et al. (2010) suggested in their model study that emissions of CH₄ and
28 N₂O from terrestrial ecosystems may offset about two thirds of the land CO₂ sink over the
29 North American continent. However, DOC fluxes were not included in these studies. Gielen
30 et al. (2011) reported that the lateral export of DOC accounted for 11% of the NEP in forests.
31 Thus, these previous studies indicate substantial contributions from non-CO₂ carbon and

1 GHG fluxes and the need for including these exchanges when estimating and/or modeling full
2 forest C and GHG budgets.

3

4 In this study, we estimated the C and GHG balances of four afforested pine stands ranging
5 from 7 to 70 years of age. The main objectives were i) to determine the effects of stand age
6 and site quality on the forest C and GHG balances and ii) to investigate the contribution of
7 non-CO₂ fluxes (i.e., CH₄, N₂O and DOC) to the forest C and GHG balances.

8

9 **2 Material and methods**

10 **2.1 Study sites**

11 This study was conducted in four eastern white pine (*Pinus strobus* L.) forests, which are part
12 of the Turkey Point Flux Station located on the north shore of Lake Erie in Southern Ontario,
13 Canada (42° 71' N, 80° 36' W). The region has a temperate climate with a 30-year mean
14 annual temperature of 8.0 °C and an annual precipitation of 1036 mm (Environment Canada
15 Norms from 1981-2010 at Delhi, Ontario). The two older forests were planted in 1939 (TP39;
16 70 years old in 2009) and 1974 (TP74; 35 years old in 2009) on cleared oak-savannah land,
17 while the two younger stands were established on former agricultural lands in 1989 (TP89; 20
18 years old in 2009) and in 2002 (TP02; 7 years old in 2009). At the oldest site TP39, a
19 moderate thinning was carried out in 1983. Although all four stands grow on generally similar
20 sandy and dry soils, the two youngest sites TP02 and TP89 are characterized by greater site
21 quality compared to the two older sites (Table 1). The higher site quality at the two younger
22 stands might result from post-agricultural remnants of nutrients (e.g. elevated soil nitrogen
23 and pH, see Table 1). In addition, high site productivity at the 20-year-old site, TP89, is
24 further due to a shallow ground water table that allows trees to have continuous access to
25 water (Peichl et al., 2010a). Each site has a tower instrumented for eddy covariance flux and
26 meteorological measurements. A suite of standard instruments for soil temperature and soil
27 moisture measurements at several depths is also installed at each site. More details of site-
28 specific instrumentation, stand and soil characteristics are given in previous studies (Arain
29 and Restrepo-Coupe, 2005; Khomik et al., 2010; McLaren et al., 2008; Peichl and Arain,
30 2006; Peichl et al., 2010a).

31

1 2.2 Biometric estimates of C pools and fluxes

2 Above- and belowground C pools in forest biomass and soil were determined in permanent
3 sample plots established at each site in 2004 following the National Forest Inventory Protocol
4 (NFI, 2003) as described in detail in Peichl and Arain (2006) and Peichl et al. (2010c). Using
5 data from these NFI inventory plots, biomass and detritus pools of canopy and understorey
6 trees, ground vegetation, and forest floor (LFH-layer) were determined each year in autumn.
7 Forest floor woody debris pools were determined using the line intersect method (Van
8 Wagner, 1968). Soil C and fine root biomass pools were determined from soil coring in 2004.
9 Litter-fall was collected using litter traps bi-weekly to seasonally (i.e. every three months) in
10 2004-2006 and bi-annually in 2007-2008 (once immediately after the peak litter fall period in
11 autumn capturing ~80% of the annual litter-fall and once in spring after the snowmelt) (Peichl
12 et al. 2010c). Forest floor litter decomposition rates were determined over four years (from
13 autumn 2004 to autumn 2008) at the three older sites. Litter bags with 1 mm mesh size were
14 filled with 10 g of air-dried pine needles and 20 bags were retrieved annually to determine the
15 mass loss.

16

17 Net primary production (NPP) was determined from the annual changes in living biomass (Δ
18 B_L) and detritus (ΔB_D) pools based on annual inventory and litter-fall data combined with
19 site-specific allometric biomass equations (Peichl and Arain, 2007) (Eq. 1). Soil respiration
20 (R_S) was measured from 2004-2007 using a LiCor LI-6400 portable chamber system (Khomik
21 et al., 2006, 2009). Soil heterotrophic respiration (RH_S) was computed using a site-specific
22 soil respiration model derived from trenched plot respiration data and further partitioned into
23 heterotrophic respiration from mineral soil (RH_M) and litter layer (RH_{LFH}) (Eq. 2) (Peichl et
24 al., 2010b, 2010c). Heterotrophic respiration from aboveground woody debris (RH_{WD}) was
25 derived from woody debris decomposition rates (Black et al., 2007; Law et al., 2001) and
26 woody debris pools and added to RH_S to obtain total heterotrophic respiration (RH) (Eq. 3).
27 Autotrophic root respiration (RA_R) was determined from the difference between R_S and RH_S
28 (Eq. 4), while autotrophic aboveground canopy respiration (RA_C) was derived by subtracting
29 RA_R from RA (Eq. 5), with RA being the difference between biometric estimates of NPP and
30 GPP (Peichl et al., 2010c). The biometric estimate of GPP was estimated from NPP assuming
31 that NPP is a constant fraction (47%) of gross primary production (GPP) on the annual scale

1 as suggested for coniferous forests by Waring et al. (1998). The biometric estimate of RE was
2 calculated as the sum of the individual respiration terms (Eq. 6).

3

$$4 \quad NPP = \Delta B_L + \Delta B_D \quad \text{Eq. (1)}$$

$$5 \quad RH_S = RH_M + RH_{LFH} \quad \text{Eq. (2)}$$

$$6 \quad RH = RH_S + RH_{WD} \quad \text{Eq. (3)}$$

$$7 \quad RA_R = R_S - RH_S \quad \text{Eq. (4)}$$

$$8 \quad RA_C = RA - RA_R \quad \text{Eq. (5)}$$

$$9 \quad RE = RH_S + RH_{WD} + RA_R + RA_C \quad \text{Eq. (6)}$$

10 The biometric estimates of individual component fluxes were used to demonstrate their
11 within-ecosystem partitioning, whereas total RE and GPP were further combined with EC
12 derived estimates on the stand level as described further below (section 2.4).

13

14 **2.3 DOC export**

15 The DOC export as the difference between input via precipitation and soil leaching below 1 m
16 depth was estimated for the snow free periods (April to December) of 2004 and 2005.
17 Precipitation was collected in plastic buckets equipped with a funnel on top of meteorological
18 towers at bi-weekly to monthly intervals and analyzed for its DOC concentration using a
19 Shimadzu 5050 Analyzer. Soil samples from 1 m depth were taken to estimate the null-point
20 DOC concentration (DOC_{np}) as indicator of the potential for DOC concentrations leaching
21 below this depth (Moore et al., 1992). The export of DOC via soil water leaching was
22 calculated by multiplying seasonal water leaching rates with DOC_{np} concentration at 1 m
23 depth. The water leaching rate was estimated as the difference between precipitation and
24 evapotranspiration (based on EC measurements) during the frost free period, assuming zero
25 run-off at these flat and sandy sites. A more detailed description is provided by Peichl et al.
26 (2007).

27

28 **2.4 Measurements of GHG (CO_2 , CH_4 and N_2O) fluxes**

1 *CO₂ fluxes*

2 Annual gross primary production (GPP), ecosystem respiration (RE), and net ecosystem
3 production (NEP) were estimated from eddy-covariance (EC) measurements at all four sites
4 from 2004 to 2008. Instrument set up and data processing procedure are described in detail in
5 Arain and Restrepo (2005) and Peichl et al. (2010a). Briefly, a closed-path eddy covariance
6 system (infra-red gas analyzer (IRGA), model LI-7000, LI-COR Inc.; sonic anemometer
7 model CSAT-3, Campbell Scientific Inc.(CSI; fine-wire thermocouple) was operated at 70-
8 year-old stand, TP39, while an open-path system (IRGA model LI-7500; LI-COR Inc.; sonic
9 anemometer model CSAT-3, CSI; fine-wire thermocouple) was rotated at bi-weekly to
10 monthly intervals among the three younger sites from 2004 to 2007. In 2008, measurements
11 were terminated at 20-year-old stand, TP89, while continuous measurements were started at
12 the 35 and 7-year-old (i.e. TP74 and TP02) sites using closed-path EC systems, comprising
13 Li-7000 IRGAs and CSAT3 sonic anemometers. For each of the three younger sites, data
14 from all years were pooled and site-specific models were developed based on
15 parameterization to environmental variables to fill the missing flux data (Peichl et al., 2010a).
16 Since the absolute values for the EC estimates of NEP, GPP and RE differed from their
17 respective biometric estimates, their means from both EC and biometric estimates are
18 presented to obtain a more robust estimate that is constrained by both methods. The mean RE
19 and GPP fluxes therefore do not exactly match the sum of their individual biometric
20 component flux estimates.

21

22 *CH₄ and N₂O fluxes*

23 Fluxes of CH₄ and N₂O were determined at monthly intervals at all four sites from April to
24 December of 2006 and 2007 using the static chambers as described by Peichl et al. (2010b).
25 Briefly, air samples (20 mL) were withdrawn using syringes at 0, 30, 60, and 90 minutes
26 through a plastic tube inserted into evacuated glass vials (13 mL) equipped with a gray butyl
27 septum in the field and analyzed within one week for CH₄ concentrations using a Shimadzu
28 Mini Gas Chromatograph (GC) (Shimadzu Scientific Instruments, Columbia, ML, USA)
29 equipped with a methanizer and a Flame Ionization Detector (FID) detector and for N₂O
30 concentrations using a Shimadzu 14-A GC (Shimadzu Scientific Instruments, Columbia, ML,
31 USA) equipped with an electron capture detector (ECD). Fluxes were calculated from the
32 linear increase in gas concentrations over the sampling period. To obtain cumulative sums,

1 fluxes were linearly interpolated and converted to CO₂ equivalent (CO₂ eq) using the global
2 warming potential (GWP, over a 100-year timeframe including carbon-climate feedbacks) of
3 34 and 298 for CH₄ and N₂O, respectively (IPCC, 2013). The mean of the two measured years
4 was combined with the 4-year mean C balance (i.e. NEP minus DOC export) in CO₂ eq to
5 obtain an estimate of the total GWP. Exchanges of CH₄, N₂O, and DOC were not estimated
6 during the snow covered months (January to March) which might have led to underestimation
7 of the total annual exchange. Especially in regions with discontinuous snow cover and
8 frequent freeze-thaw events, large winter emissions of N₂O may occur during these periods
9 (Luo et al., 2012; Teepe et al., 2001). However, winter season fluxes of CH₄, N₂O, and DOC
10 in forests that experience severe winters with continuous snow cover are generally small
11 (Ågren et al., 2007; van Bochove et al., 2000; Yashiro et al., 2006). Since our study sites
12 experience temperatures below freezing and continuous snow cover from December to
13 March, these winter fluxes were assumed to have a negligible effect on the total C and GHG
14 balances in our study.

15

16 **2.5 Site quality effects on cumulative NEP**

17 To estimate the total C sequestration from the cumulative NEP over the entire age-sequence,
18 measured annual NEPs from the four sites were linearly interpolated to obtain a 70-year-
19 record of annual NEP. Following Peichl et al. (2010a), two scenarios were developed in
20 which NEP of four sites was normalized by differences in their site indices (SI). The SI is a
21 common measure to describe the site quality and is used as predictor for tree growth in forest
22 stands (e.g. Milner, 1992). In the first ‘low productive’ scenario, it was assumed that all four
23 stands grow on low quality sites by applying a SI correction on NEP for TP02 and TP89 to
24 match the SI of TP74 and TP39. Conversely, in the second ‘high productive’ scenario, NEP at
25 TP74 and TP39 was normalized to match the SI of TP02 and TP89 following the assumption
26 that all four stands grow on high quality sites.

27

28 **3 Results**

29 **3.1 Forest C pools and fluxes**

30 Needle litter decomposition rates did not differ significantly among the three older sites with
31 the exponential decay coefficient k ranging from 0.28 to 0.31 (Figure 1). The rates of needle

1 litter-fall exceeded those of decomposition resulting in a net accumulation of 3.7, 2.0 and 3.2 t
2 $\text{ha}^{-1} \text{y}^{-1}$ at the 20, 35 and 70-year-old stands, respectively (Figure 2).

3

4 The total ecosystem C pool (including vegetation and soil) increased with age from 46 t C ha^{-1}
5 at the youngest site to 197 t C ha^{-1} at the oldest site (Figure 3). The largest age-related
6 differences in C pools among sites occurred in stem and root biomass, whereas the canopy
7 (foliage and branches) C pool was similar among the three older stands. The sum of the
8 secondary C pools, such as woody debris, understorey and ground vegetation, were small and
9 similar among three younger sites (ranging from 1.5 to 2.7 t C ha^{-1}) compared to that of the
10 70-year-old stand (11.3 t C ha^{-1}).

11

12 The NPP ranged between 379 g C $\text{m}^{-2} \text{y}^{-1}$ at the 7-year-old stand to 835 g C $\text{m}^{-2} \text{y}^{-1}$ at the 20-
13 year-old stand. Age-related differences in RE were mainly driven by changes in RA and RH
14 from aboveground biomass and litter, whereas belowground root respiration and soil RH were
15 similar among the four stands.

16

17 **3.2 Forest C balance**

18 The mean NEP over the four years was 70, 590, 280 and 130 g C $\text{m}^{-2} \text{y}^{-1}$ at the 7, 20, 35 and
19 70-year-old stands, respectively (Figure 3). The 2-year mean DOC export decreased with
20 stand age from 7 g $\text{m}^{-2} \text{y}^{-1}$ at the 7-year-old stand to 4, 3 and 2 g $\text{m}^{-2} \text{y}^{-1}$ at the 20, 35 and 70-
21 year-old stands, respectively (Figure 3). The relative contribution of the net DOC export (i.e.
22 the difference between DOC input via precipitation and DOC export at 1 m depth) to NEP
23 decreased from 10% at 7-year-old stand to less than 1-2% at the three older stands (Figure
24 4a).

25

26 **3.3 Forest GHG balance**

27 Across the four stands, the combined N_2O and CH_4 flux ranged from -0.4 to 0.1 t $\text{CO}_2 \text{eq ha}^{-1}$
28 y^{-1} during the two measurement years (Figure 3). The net GHG balance derived from the
29 mean C balance and the combined N_2O and CH_4 flux was -2.6, -21.6, -13.5 and -4.8 t $\text{CO}_2 \text{eq}$
30 $\text{ha}^{-1} \text{y}^{-1}$ at the 7, 20, 35 and 70-year-old stands, respectively (Figure 3). The combined relative

1 contribution of CH₄ and N₂O to the GHG balance ranged from <1% at the 20 and 35-year-old
2 stands to 13% and 8% at the 7 and 70-year-old stands, respectively (Figure 4b).

3

4 **3.4 Site quality effect on the cumulative NEP**

5 Over the initial 70 years following afforestation, the cumulative CO₂ sequestration ranged
6 from 129 t C ha⁻¹ for a low-productive (i.e. site index = 26) forest to 297 t C ha⁻¹ for highly
7 productive (i.e. site index = 60) forest (Figure 5). The C compensation point (i.e. the timing
8 when net CO₂ accumulation becomes positive) was reached about 10 years after plantation
9 establishment in both scenarios.

10

11 **4 Discussion**

12 **4.1 The forest C balance across stand age and site quality**

13 Following afforestation of low productive agricultural and other marginal land, the balance of
14 needle litter production and decomposition is a major determinant of the accumulation and
15 partitioning of organic matter into soil organic carbon accumulation and losses due to
16 heterotrophic respiration (Yang et al., 2011). In our study, we did not find any age-related
17 differences for organic matter turnover when comparing litter-fall and decomposition rates
18 among the three older sites. Decomposition rates were within the range of those previously
19 reported for other Canadian coniferous forests (Moore et al., 1999; Trofymow et al., 2002)
20 and appeared to be unaffected by either stand age or site quality. In contrast, age-related
21 differences in the litter-fall rates were apparent when comparing the 35 and 70-year-old
22 stands, with the latter one having higher rates (but similar SI values). However, across the
23 entire age-sequence, an age effect on litter-fall previously observed in other studies (e.g. Law
24 et al., 2001) was likely masked by the high litter-fall rates at our 20-year-old high productive
25 site.

26

27 In contrast to the small differences in organic matter turnover, we observed manifold changes
28 in the magnitudes and partitioning of the C fluxes among the four stands. These changes were
29 primarily driven by the differences in aboveground biomass and flux components. It is further
30 noteworthy that the changes in individual biomass pools did not always reflect the changes in

1 their associated in- and/or outgoing C fluxes. For instance, a small difference in RA_R despite
2 manifold changes in fine root biomass among the four stands was observed. This might have
3 resulted from i) a temporal mismatch in measurements (fine root biomass was determined in
4 2004 when seedling trees at the youngest site were only 2 years old whereas root respiration
5 was also estimated during the subsequent years (2004-2008) of rapid tree seedling and
6 herbaceous ground cover development), ii) a masking effect from greater understorey and/or
7 groundcover root respiration at the youngest and oldest stands and iii) contrasting patterns in
8 the allocation of assimilates from the canopy to the roots in the high- *versus* low-productive
9 stands (Vicca et al., 2012). Thus, this observation indicates some limitations to inferring C
10 fluxes solely from the magnitude of the biomass pools.

11
12 Although stand age and site quality were not fully replicated among the four sites, we suggest
13 that, given the similar climatic conditions, most of the observed differences in the C balances
14 might be explained by either one or both factors. Age-related differences become most
15 evident when comparing the two younger (both having a similar high SI) and the two older
16 sites (both having a similar low SI), respectively. In both cases, the comparison suggests
17 greater NEP and NPP in the middle age stands (20 and 35-year-old) compared to the young
18 and mature stand, respectively. An additional effect of site quality may explain the large
19 difference in NEP and NPP between the two middle-age stands, with considerably higher
20 values noted at the 20-year-old stand characterized by a higher (i.e. double) SI value. In
21 contrast to NEP and NPP, mean GPP and RE increased from the 7 to the 20-year-old stand as
22 well as from the 35 to 70-year-old stand. This indicates that GPP and RE have not yet reached
23 their maxima in this 70-year-old age sequence. Higher GPP and RE at the 20-year-old site,
24 compared to the two older sites, were likely due to the additional effect of higher site quality
25 due to greater water availability at this site (Peichl et al., 2010a). These results based on the
26 mean values for NEP, GPP and RE from biometric and EC estimates agree with those from
27 using estimates from either one of these two methods only since their age-related patterns
28 among sites are generally similar although their absolute values differ between 9 and 56%. A
29 detailed discussion on the underlying reasons for the deviation between the two methods was
30 previously provided by Peichl et al. (2010c). Similar age patterns for forest production and
31 respiration were reported for other forest age-sequences (Goulden et al., 2011; Law et al.,
32 2003; Luyssaert et al., 2007; Pregitzer and Euskirchen, 2004). However, in contrast to

1 naturally regenerating forests, the switch from source to sink and the peak in NEP and NPP
2 occurred about 10-20 years earlier in our afforested plantation stands, which highlights their
3 potential for rapid C sequestration.

4

5 The contribution of DOC to NEP was considerable at the youngest site (10%), while it was
6 rather low (<2%) at the three older sites. Previous work at these sites suggested that the
7 reduced DOC export resulted from both a decrease in DOC concentrations at 1 m soil depth
8 with increasing stand age as well as from decreased water leaching in the older stands
9 compared to the youngest stand (Peichl et al., 2007). In agreement with our study, Kindler et
10 al. (2011) found that DOC leaching hardly affected NEP at the majority of the forests
11 investigated in their study. A decrease in soil solution leaching due to greater canopy
12 interception and root water uptake and/or the increase in adsorption of DOC to soil particles
13 due accumulation of Fe and Al with stand age might explain the reduced DOC export in older
14 forest stands (Camino-Serrano et al., 2014; Kothawala et al., 2009; Peichl et al., 2007). In
15 contrast, a higher contribution of DOC of 11% was observed in a 80-year-old Belgium Scots
16 pine stand (Gielen et al., 2011). Thus, depending on site productivity and hydrology, the
17 relative contribution of the DOC export to the forest C balance might vary across forest
18 ecosystems and become significant also at some of the mature forest sites.

19

20 The accumulated C sequestration simulated over the entire 70 years of the age-sequence was
21 substantial in both the high and low productive scenarios. Moreover, the difference (by a
22 factor of 2) due to site quality was considerable. The importance of site quality effects on
23 forest growth is widely recognized in traditional forest research (McLeod and Running, 1988;
24 Milner, 1992; Pietrzykowski, 2014; Vose and Allen, 1988) as well as in recent studies on the
25 C allocation within forest stands (e.g. Vicca et al., 2012). Our study further highlights the
26 need for accounting for differences in site quality when assessing forest C and GHG balances
27 across forest ecosystems and to improve their up-scaling beyond ecosystem boundaries.

28

29 **4.2 Forest GHG balance across stand age and site productivity**

30 To date, few attempts have been made to quantify the total forest GHG balance and the
31 relative contribution of individual components by including all relevant fluxes (i.e. CO₂, CH₄,

1 N₂O and DOC) (Ball et al., 2007; Luysaert et al., 2012; Schulze et al., 2009; Zona et al.,
2 2013). Previous studies estimated GHG balances for individual forests at about +3 t CO₂ eq
3 ha⁻¹ y⁻¹ for a young short-rotation poplar plantation (Zona et al., 2013) and approximately -15
4 t CO₂ eq ha⁻¹ y⁻¹ for a middle-age pine forest (Ball et al., 2007), respectively, which is close to
5 the range observed in our study. Both of these studies also agree with our findings of non-CO₂
6 GHGs contributing considerably to the GHG balance in recently established plantations (as
7 well as in the mature 70-year-old forest in our study), whereas the CO₂ exchange dominates
8 the GHG balance in middle-aged forests. Similarly, Inatomi et al. (2010), based on results
9 from a process-based model, found that the GHG balance was mainly driven by the CO₂
10 exchange in a 50-year-old cool-temperate deciduous forest. However, the contribution of non-
11 CO₂ GHG could be substantial in poorly-drained locations within temperate forests (Ullah
12 and Moore, 2011). The relative contribution of CH₄ and N₂O to the forest GHG balance might
13 further be considerably affected by more frequent freeze-thaw events (Luo et al., 2012; Teepe
14 et al., 2001), altered N input (Liu and Greaver, 2009) as well as by contrasting forest
15 management and tree growth responses to climatic changes in the future (Metsaranta et al.,
16 2011; Ximenes et al., 2012). For instance, Metsaranta et al. (2011) suggested that the
17 cumulative GWP over 70 years for a coniferous forest in British Columbia may vary between
18 -67 and 67 t CO₂ eq ha⁻¹ in the best and worst-case modeling scenario. Thus, the manifold
19 variation in the magnitude of GWP and the relative contribution of non-CO₂ gases to the
20 GWP in these studies and among the four stands in our study emphasizes that the forest GHG
21 balance may vary widely within a heterogeneous (i.e. in terms of age and site quality) forest
22 landscape, which needs to be considered when extrapolating findings from individual forest
23 stands to regions.

24

25 Overall, our study advances the current understanding of the forest GHG balance by
26 demonstrating that the magnitude and contribution of individual GHGs may have manifold
27 variations in forests due to differences in stand age and site quality. Both factors determine
28 the forest NEP and thus the magnitude of the CO₂ flux while in comparison having a
29 relatively small effect on the CH₄, N₂O and DOC fluxes. Thus, our findings suggest a link
30 between NEP and the relative contribution of individual gases to the GHG balance with the
31 implication that the ecosystem exchange of CO₂ is the dominant driver of the forest GHG
32 balance partitioning. As a consequence, the contribution of CH₄, N₂O and DOC fluxes to the

1 GHG balance might be low in highly productive (e.g. middle-age) forests whereas it may be
2 more important in low productive (e.g. recently established and mature) forests due to the
3 differences in NEP (as opposed to changes in the non-CO₂ fluxes). Understanding the changes
4 in individual contribution of forest C and GHG exchanges to the total GWP throughout their
5 life cycle is imperative to evaluate the potential of these ecosystems as a tool in mitigating
6 global warming and the increase of atmospheric GHG concentrations.

7

8 **5 Conclusions**

9 We combined C pools and fluxes of CO₂, CH₄, N₂O and DOC to estimate C and GHG
10 balances for an age-sequence (spanning 7 to 70 years) of afforested white pine stands in the
11 temperate region of southern Ontario, Canada. Based on our findings we conclude that:

- 12 • The magnitudes and within-stand partitioning of CO₂ fluxes were highly variable with
13 stand age and site quality.
- 14 • The role of DOC export for the C balance was substantial in the recently established pine
15 plantation, but small to marginal in the highly productive maturing stands.
- 16 • The combined exchanges of CH₄ and N₂O significantly contributed to the GHG balance of
17 the young and mature pine forests due to their lower NEP.
- 18 • Stand age and site quality may cause manifold differences in the forest C and GHG
19 balances and need to be accounted for to improve their up-scaling from ecosystems to
20 regions.
- 21 • Overall, these temperate pine afforestation stands act as a substantial C sink and provide a
22 considerable negative GWP throughout the maturing phase.

23

24

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11

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1 Table 1: Site characteristics of the four Turkey Point pine forest stands (adopted from Peichl
 2 et al. (2010a)).

Site code	TP02	TP89	TP74	TP39
Planting year	2002	1989	1974	1939
Age in 2009 (years)	7	20	35	70
Tree height (m) [*]	3.6 ± 0.4	13.4 ± 0.9	13.5 ± 0.7	22.5 ± 1.4
Tree diameter at breast height (cm) [*]	5.3 ± 0.9	18.2 ± 0.6	16.7 ± 0.3	35.9 ± 5.9
Stem density (stems ha ⁻¹) [*]	1683 ± 189	1317 ± 251	1608 ± 138	421 ± 166
Leaf Area Index (m ² m ⁻²) ^{**}	1.5	12.8	5.6	8.0
Site index (SI ₂₅) ^{***}	60	55	28	26
Forest floor C:N ratio	N.A.	16.1	24.5	17.4
Mineral soil C:N ratio (0-10 cm)	11.4	14.2	19.4	15.4
Soil N (0-10 cm) (g m ⁻²)	86	94	67	67
Soil C (0-55 cm) (g m ⁻²)	3720	3390	3670	3670
Soil pH _(CaCl) (0-10 cm)	6.3	4.3	3.7	4.1
Mean water table depth (m)	2-3.5	0.5-2	6-7	6-7

3 ^{*} in 2008

4 ^{**} Chen et al. (2006)

5 N.A. = not available

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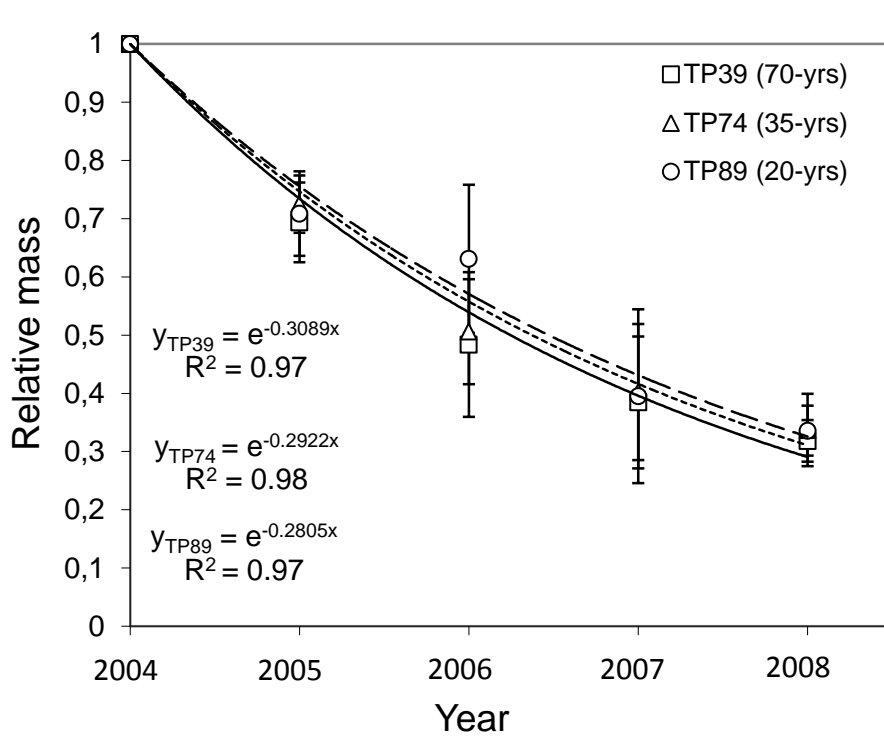
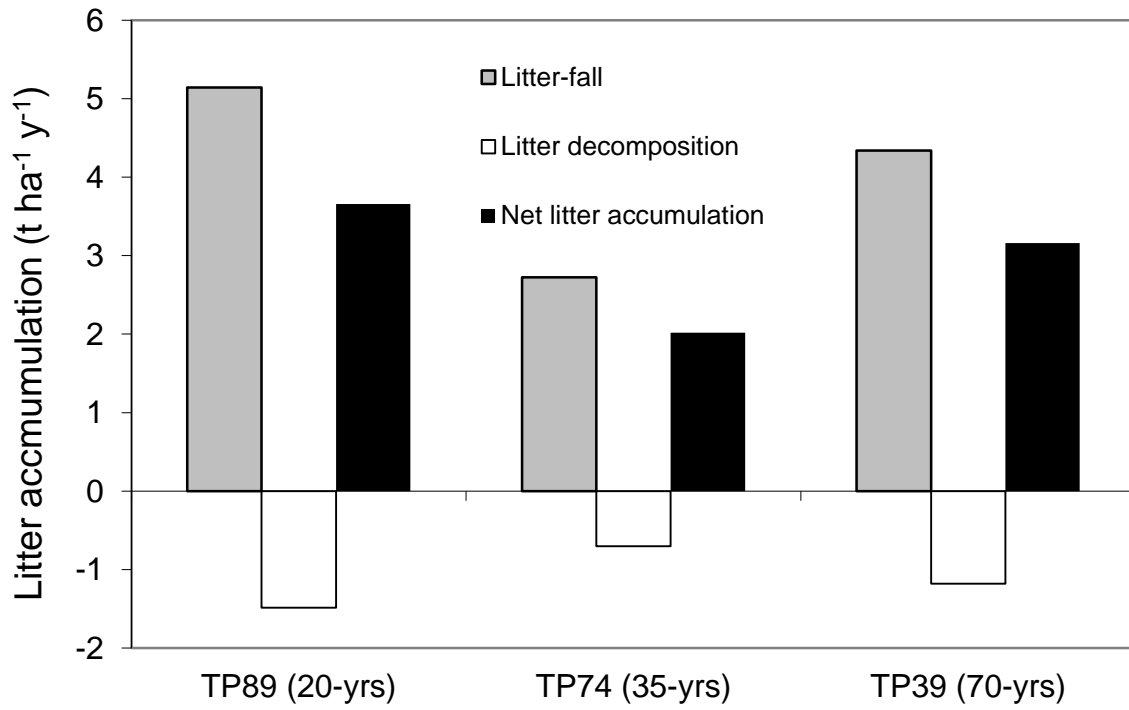
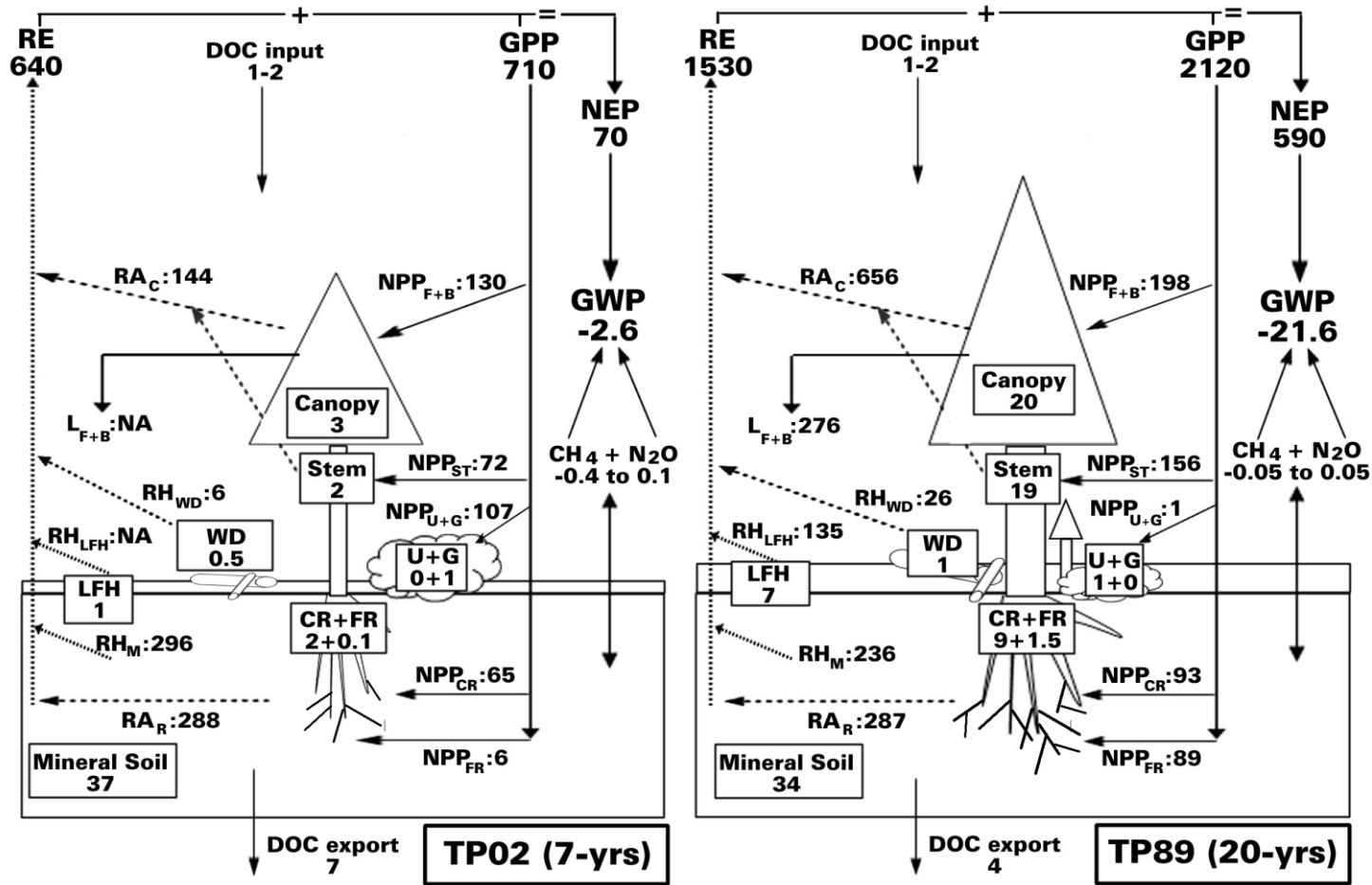


Figure 1: Needle litter decomposition rates over four years (2004-2008) at the three older Turkey Point pine forest sites TP89, TP74 and TP39 (20, 35 and 70 years old, respectively).



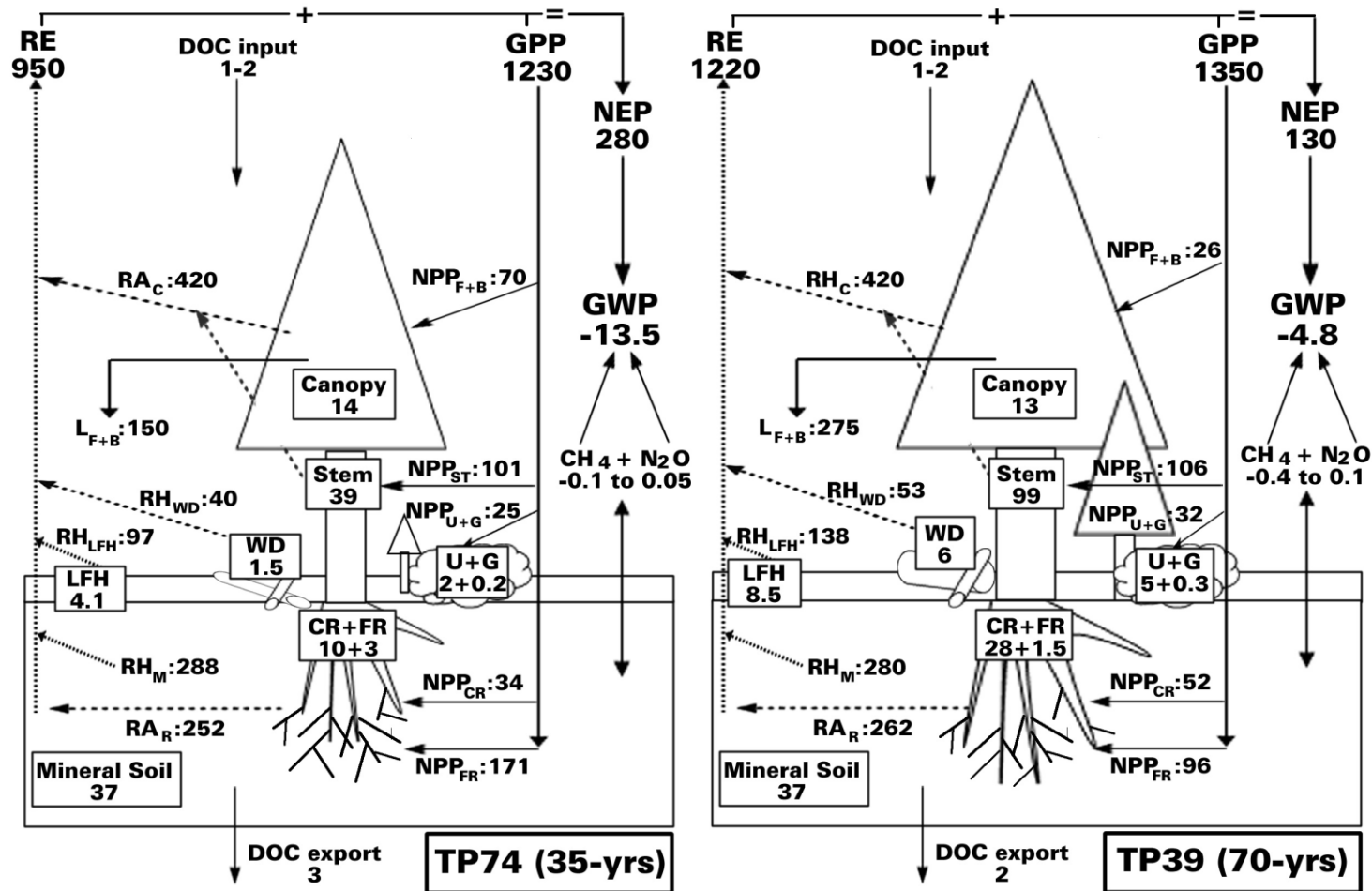
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 2 Figure 2: Needle litter-fall, decomposition and net accumulation on the forest floor over four
 3 years (2004-2008) at the three older Turkey Point pine forest sites TP89, TP74 and TP39.

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2 **Figure 3:** Ecosystem carbon (C) pools (square boxes; t C ha⁻¹), C fluxes (gross and net primary production (GPP, NPP), litter-fall and DOC flux
 3 = solid arrows, ecosystem respiration (RE) fluxes = dotted arrows; g C m⁻² y⁻¹), CH₄ and N₂O fluxes (t CO₂ eq ha⁻¹ y⁻¹) and GHG balance
 4 expressed as global warming potential (GWP; t CO₂ eq ha⁻¹ y⁻¹) at the four Turkey Point pine forest sites, TP02, TP89, TP74 and TP39.



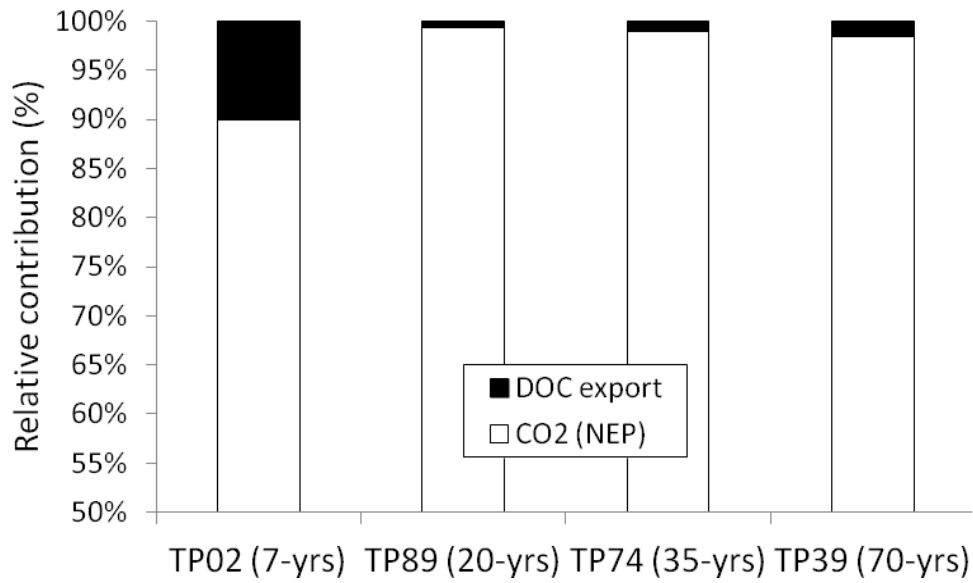
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2 Figure 3 continued

3 Abbreviations: C = canopy; F = foliage; B = branches; ST = stem; LFH = forest floor; WD = woody debris; CR = coarse roots; FR = fine roots; U = understorey; G = ground

4 vegetation; M = mineral soil; R = roots; RH = heterotrophic respiration; RA = autotrophic respiration; L = litter-fall; DOC = dissolved organic carbon; NA = not available

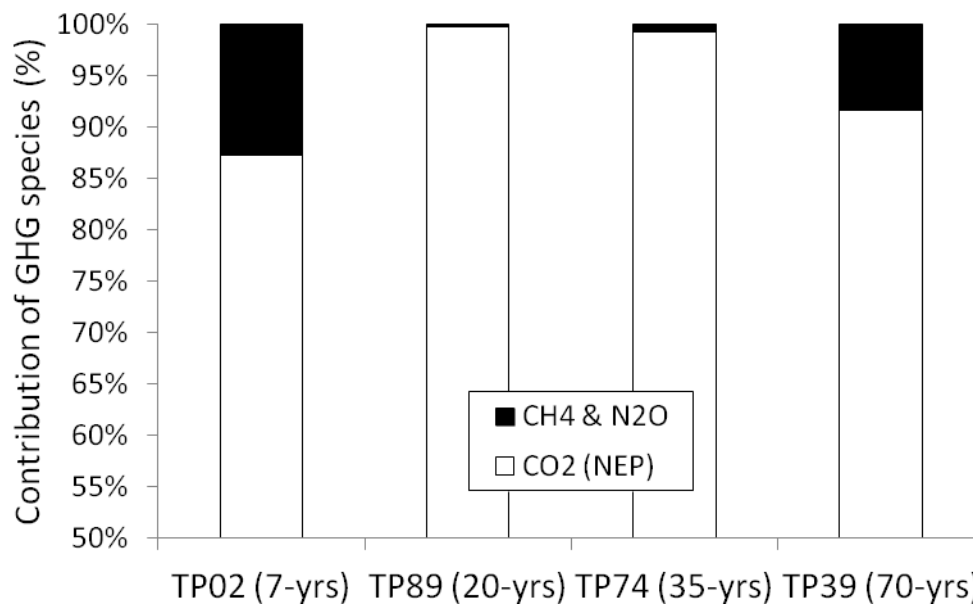
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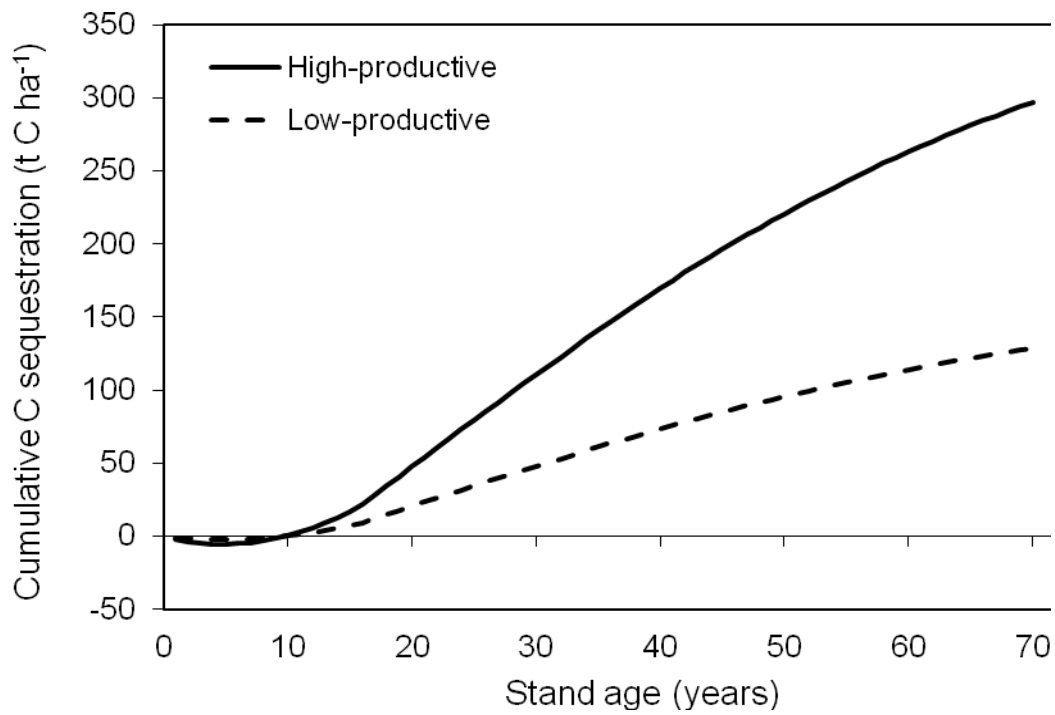
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6 Figure 4: (a) Relative contributions of the DOC export (below 1m depth) and NEP to the total
7 C balance and (b) relative contribution of the CO₂ exchange and the combined CH₄ and N₂O
8 exchange to the GHG balance at the four Turkey Point pine forest sites, TP02, TP74, TP89
9 and TP39.

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Figure 5: Cumulative C sequestration assuming high (site index = 60; solid line) and low (site index = 26; dashed line) site quality for all four Turkey Point pine stands.