



Consequences of long-term severe industrial pollution for aboveground carbon and nitrogen pools in northern taiga forests at local and regional scales



Sirkku Manninen ^{a,*}, Vitali Zverev ^b, Igor Bergman ^c, Mikhail V. Kozlov ^b

^a Department of Environmental Sciences, University of Helsinki, P.O. Box 65 (Viikinkaari 2a), FI-00014 Helsinki, Finland

^b Section of Ecology, Department of Biology, University of Turku, FI-20014 Turku, Finland

^c Institute of Plant and Animal Ecology, Ural Branch, Russian Academy of Sciences, ul. Vos'mogo Marta 202, Yekaterinburg 620144, Russia

HIGHLIGHTS

- Pollution changed the carbon and nitrogen concentrations in plant tissues.
- Plant biomass near the smelter was 1% of that in unpolluted forests.
- Pollution reduced plant biomass over an area of about 107,200 km².
- The regional loss of phytomass carbon stock was estimated at 4.24×10^{13} g C.
- Regional carbon stock was more affected by pollution than by fire and insect pests.

GRAPHICAL ABSTRACT



Deteriorated ecosystem that developed from a spruce forest under chronic pollution exposure (8 km south of the Monchegorsk smelter, Kola Peninsula, north-western Russia). Photo: V. Zverev.

ARTICLE INFO

Article history:

Received 27 April 2015

Received in revised form 20 July 2015

Accepted 21 July 2015

Available online xxxx

Editor: D. Barcelo

Keywords:

Boreal forest

Carbon sequestration

Monchegorsk

Nickel–copper smelter

Nitrogen concentration

Phytomass

ABSTRACT

Boreal coniferous forests act as an important sink for atmospheric carbon dioxide. The overall tree carbon (C) sink in the forests of Europe has increased during the past decades, especially due to management and elevated nitrogen (N) deposition; however, industrial atmospheric pollution, primarily sulphur dioxide and heavy metals, still negatively affect forest biomass production at different spatial scales. We report local and regional changes in forest aboveground biomass, C and N concentrations in plant tissues, and C and N pools caused by long-term atmospheric emissions from a large point source, the nickel–copper smelter in Monchegorsk, in north-western Russia. An increase in pollution load (assessed as Cu concentration in forest litter) caused C to increase in foliage but C remained unchanged in wood, while N decreased in foliage and increased in wood, demonstrating strong effects of pollution on resource translocation between green and woody tissues. The aboveground C and N pools were primarily governed by plant biomass, which strongly decreased with an increase in pollution load. In our study sites (located 1.6–39.7 km from the smelter) living aboveground plant biomass was 76 to 4888 g m⁻², and C and N pools ranged 35–2333 g C m⁻² and 0.5–35.1 g N m⁻², respectively. We estimate that the aboveground plant biomass is reduced due to chronic exposure to industrial air pollution over an area of about 107,200 km², and the total (aboveground and belowground) loss of phytomass C stock amounts to 4.24×10^{13} g C. Our results emphasize the need to account for the overall impact of industrial polluters on ecosystem C and N pools when

* Corresponding author.

E-mail address: sirkku.manninen@helsinki.fi (S. Manninen).

assessing the C and N dynamics in northern boreal forests because of the marked long-term negative effects of their emissions on structure and productivity of plant communities.

© 2015 Elsevier B.V. All rights reserved.

1. Introduction

Annual global carbon dioxide (CO₂) emissions from fossil fuel combustion and industry reached $9.5 \pm 0.5 \times 10^{15}$ g C in 2011 (Le Quéré et al., 2013), stressing the need to increase the carbon (C) sequestration capacity of ecosystems. Boreal coniferous forests cover an area of 1.14×10^7 km² and are an important C storage globally. They provided a C sink of $5.0 \pm 1.0 \times 10^{14}$ g C year⁻¹ in 1990–2007 (Pan et al., 2011); however, their C pools and dynamics are still insufficiently understood (Lorenz and Lal, 2010; and references therein). The overall stability of the forest C sink is the net result of contrasting C dynamics in different countries and regions associated with the climate, soil fertility, natural disturbances and forest management, as well as atmospheric pollution. For example, the boreal forests in European Russia and northern Europe have shown marked increases in the live vegetation biomass and C stock since the 1950s (Pan et al., 2011, and references therein), mainly due to management and increased atmospheric nitrogen (N) deposition (Graven et al., 2013; Kauppi et al., 1992; Magnani et al., 2007). In contrast, the biomass C sinks in boreal forests in Canada and Asian Russia have been reduced by intense wildfires and insect outbreaks (Goodale et al., 2002; Kurz and Apps, 1999; Pan et al., 2011).

Industrial pollution of forests has long been recognised as having serious adverse environmental effects at the local, regional and global levels (Fowler et al., 1999; Kozlov et al., 2009; Matyssek et al., 2012). For decades, ecologists have studied the pollution effects on biota in the vicinities of large industrial areas; consequently, acute local effects of pollution on forests are reasonably well documented (reviewed by Kozlov et al., 2009).

Regional effects of air pollution attracted considerable attention in the 1970s, when large areas of forests required rehabilitation to mitigate the direct impacts of sulphur dioxide (SO₂) and acidic deposition. The problem was particularly apparent in Central Europe, with the most striking example being the 'Black Triangle', an area along the German–Czech–Polish border, where industrial air pollution caused widespread declines in high elevation conifer stands (Vancura et al., 2000). In the USA, intense episodes of elevated levels of ozone (O₃), formed from the emissions of nitrogen oxides (NO_x), carbon monoxide, and volatile organic compounds from millions of cars, have been well recognised, especially in the Los Angeles Basin, since the early 1950s. The contaminated air masses move inland with the westerly on-shore winds and are pushed against the San Bernardino Mountains, damaging and even killing sensitive ponderosa and Jeffrey pines (Bytnerowicz et al., 2008). Across the conterminous USA, the elevated O₃ levels decreased the C sink to vegetation by at least 3–12% in the 1980s (Felzer et al., 2004).

Although sulphur (S) emissions have decreased markedly in Europe and North America since the 1980s (Stern, 2006), SO₂ together with particles containing heavy metals still are the major air pollutants in the vicinity of many smelters and power plants in Europe, and especially in Russia (Kozlov et al., 2009). Model calculations demonstrate that, by 2050, severe regional problems associated with pollution are likely to occur in South-Eastern Asia, South Africa, Central America and along the Atlantic coast of South America (Fowler et al., 1999), with global consequences for C cycles, primary productivity and other characteristics of forest ecosystems.

The effects of industrial S and heavy metal emissions on forest ecosystems—in terms of pollutant accumulation in organisms and changes in species composition, abundance and fitness—have been extensively studied for decades (reviewed by de Vries et al., 2014; Freedman, 1989; Kozlov et al., 2009; Kozlov and Zvereva, 2011;

Matyssek et al., 2012; Treshow, 1984). These studies indicate that industrial air pollutants disturb C and N cycling due to impairment of photosynthesis and decreases in plant growth, increases in production of C containing secondary metabolites, reduction of translocation of C to roots and impairment of root development and function (Matyssek et al., 2012; Schütt and Cowling, 1985; Treshow, 1984). However, hardly any attention has been paid to C and N pools in industrially polluted areas (but see Fischer et al., 1995), despite the importance of C and N cycling for the structure and functions of forest ecosystems. The magnitude of the effects of industrial pollution on C and N budgets and the spatial extent of these effects remain unknown.

The goal of this study was to explore effects of long-term severe industrial pollution on aboveground C and N pools in subarctic forest ecosystems using the impact zone of the Ni–Cu smelter in Monchegorsk, in north-western Russia, as an example. First, we tested the hypothesis that pollution affects C and N concentrations in aboveground plant tissues. Second, we explored the dose-dependence relationships between pollutant load, aboveground plant biomass and C and N pools in forest ecosystems of the study region. Third, we used the determined dose-dependence relationships to estimate the total losses of the C pool and C sequestration capacity caused by industrial air pollution in the forested areas of the northern Fennoscandia, i.e. the Kola Peninsula in Russia and northern Finland, Sweden and Norway.

2. Material and methods

2.1. Emissions, air quality and deposition

The Ni–Cu smelter located near Monchegorsk (67°56' N, 32°49' E; Fig. 1) was one of the largest polluters in the Northern hemisphere for decades. The smelter was started up in 1937–1938 and had no air-cleaning facilities until 1968. The annual emissions of SO₂ reached a maximum of 278 000 t in 1983, steadily declined to about 100 000 t by mid-1990s, dropped to 45 000 t in 1999 and have remained at about this level since then (Kozlov et al., 2009). In the 1980s, the annual mean SO₂ concentrations exceeded 60 µg m⁻³ at the vicinity of the smelter (Tuovinen et al., 1993), and the total S deposition ranged from 2 to 5 g m⁻² year⁻¹ at the most polluted sites, exceeding the background values by a factor of ten or more (Prank et al., 2010). Metal emissions during the 1980s–1990s amounted 3–8000 t of Ni and 1–6 000 t of Cu annually. Annual emissions of NO_x were 3–6 000 t in the 1980s, but have decreased since then to approximately 1 000 t in the 2000s (Kozlov et al., 2009).

2.2. Nature and climate in the study area

The impact zone of the Monchegorsk smelter lies 150 km south of the tree line. Virgin Scots pine (*Pinus sylvestris* L.) stands and an impenetrable Norway spruce (*Picea abies* (L.) Karst.) forest dominated the lowland vegetation in the study area prior the building of the smelter. As a result of the pollution, forests up to 6 km distance from the smelter had perished already by 1946. Observations in the early 1990s revealed forest death in an area of 400 to 500 km², and the visible injuries to conifers were detected up to 50–60 km away from the smelter; the total area affected by air pollution was estimated to exceed 10 000 km² (Kozlov et al., 2009).

The previously forested areas in the vicinity of the smelter have been transformed into industrial barrens—bleak open landscapes with small patches of vegetation surrounded by bare land (Fig. S1a). In these habitats, conifers are practically absent, and low-stature (0.2–3 m

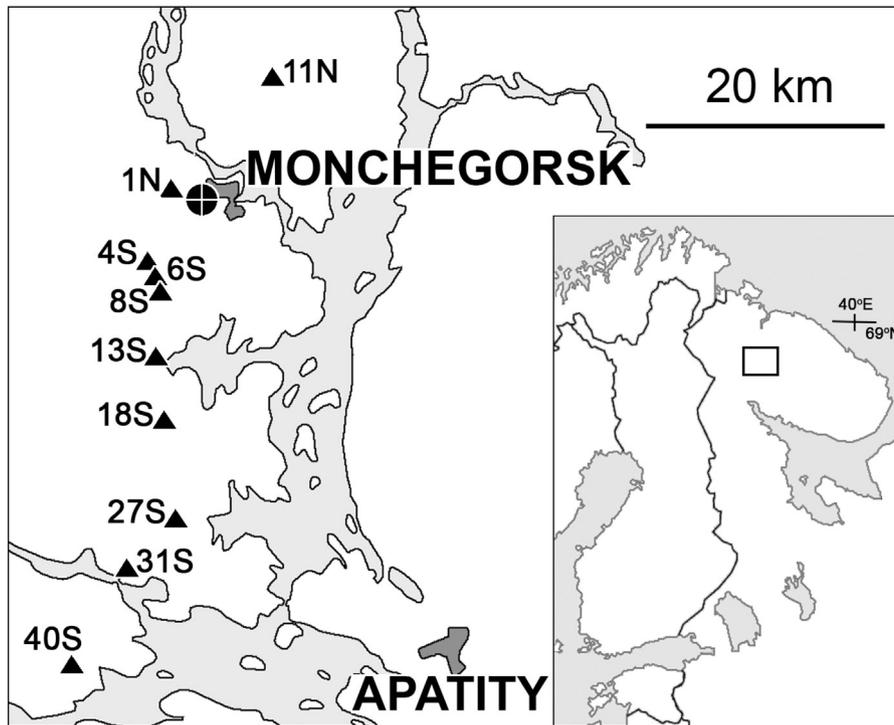


Fig. 1. Location of the study sites (triangles) in the vicinity of Monchegorsk Ni–Cu smelter, Kola Peninsula. Inserted: position of the study area in Northern Europe.

tall) mature birches growing 5–15 m apart dominate the vegetation. In the intermediate zone, the top canopy is formed by Norway spruces with visible signs of damage (dead upper canopies, low needle longevity) and mountain birches, while field- and ground-layer vegetation is sparse (Fig. S1b). In visibly unaffected Norway spruce forests, mountain birches (*Betula pubescens* ssp. *czerepanovii* (Orlova) Hämet-Ahti) are also common, and *Empetrum nigrum* ssp. *hermaphroditum* (Hagerup) Böcher, *Vaccinium myrtillus* L. and *Vaccinium vitis-idaea* L. dominate in the dense field-layer (Fig. S1c). While the conifers at the least polluted sites are reaching the age of 160–240 years, the largest alive trees at visibly damaged sites are younger than 50–75 years old.

The mean temperature in January in Monchegorsk is -13.8 °C and in July 14.1 °C, and the annual precipitation is 561 mm. The frost-free period ranges from 50 to 100 days. The cool summer lasts for ca. 3 months; the sum of effective temperatures (over $+5$ °C) is 780–800 degree-days. Northern and north-eastern winds are most frequent in summer; southern and south-western winds occur mainly in other seasons (Rigina and Kozlov, 2000).

2.3. Study sites

The data were collected at 10 sites located 1.6–39.7 km from the smelter (Fig. 1). The sites were selected for a large-scale Finnish–Russian project using the protocol described by Vorobeichik and Kozlov (2012). At the first stage, representative study sites were selected (Table 1); these, to the best of our knowledge, had similar vegetation during the pre-industrial period. Some trees may have been logged from the sites more than 50 years ago, but none of our sites have been affected by logging or fire for the last 40 years. At the second stage, 12 sampling plots 25×25 m in size were marked at each site and three of these plots were randomly selected for the study.

The level of environmental contamination was quantified by Cu concentration in forest litter (Table 1), which correlates with other pollutants, including SO_2 (Kozlov et al., 2009), and strongly decreases with the distance from the smelter ($r = 0.99$, $n = 10$, $P < 0.001$). The sampling and analytical procedures for litter chemistry were described by Eeva et al. (2012).

Table 1
Description of study sites.

Site code ^a	Latitude (N)	Longitude (E)	Altitude, m a.s.l	Distance from polluter, km	Habitat type	Litter Cu, $\mu\text{g g}^{-1}$ d.w. ^b	pH ^b
Polluter	67°55'15"	32°50'18"	140				
1 N	67°56'04"	32°58'19"	180	1.6	Industrial barren	3132	4.2
4S	67°52'59"	32°46'40"	210	4.3	Birch transitional community	2743	4.2
6S	67°51'58"	32°47'50"	260	5.7	Industrial barren	1923	3.7
8S	67°51'01"	32°48'10"	240	7.5	Industrial barren	1874	3.7
11 N	68°00'57"	32°58'08"	160	11.5	Severely damaged spruce forest	939	3.8
13S	67°48'03"	32°47'05"	140	13.0	Severely damaged spruce forest	1023	3.7
18S	67°45'31"	32°48'29"	210	17.5	Severely damaged spruce forest	997	3.6
27S	67°40'39"	32°49'27"	220	26.7	Slightly damaged spruce forest	250	3.5
31S	67°38'21"	32°45'00"	170	31.1	Slightly damaged spruce forest	170	3.7
40S	67°34'36"	32°33'03"	140	39.7	Undamaged spruce forest	107	3.8

^a The site codes indicate approximate distance from the smelter in km and direction to the north or to the south of the smelter.

^b Forest litter Cu concentration and pH for samples collected in August 2008 after Eeva et al. (2012).

2.4. Plant biomass

We were primarily interested in C sequestration capacities, and therefore collected data on living, aboveground plant biomass. The data were separately collected for (a) top-canopy trees, i.e. trees with DBH (trunk diameter at the 1.3 m height) >5 cm; (b) understory, i.e. trees with DBH <5 cm, and shrubs; (c) field-layer vegetation; and (d) mosses and lichens. In July 2009, DBH and height of all top-canopy trees in the study plots were measured, as were basal diameter and height of all understory trees and shrubs (separately for each stem in multi-stem shrubs). After measurements, representative samples of understory plants were collected, leaves/needles were separated from woody parts, and the samples were dried at 105 °C for 48 h (leaves/needles) or for 96 h (woody parts) and weighed to the nearest 0.1 g.

The biomass of top-canopy trees was calculated (separately for foliage and for woody parts) from DBH and height using regression equations provided in earlier publications (Table S1). The biomass of understory trees and shrubs was calculated from species-specific regression equations based on the analysis of the collected specimens (Table S1).

The biomass of field-layer vegetation was measured in five 0.5 × 0.5 m subplots, randomly selected within each 25 × 25 m plot. All plants within each subplot were cut at the ground level, sorted by species and then by fractions (foliage, fruits, woody parts), dried at 105 °C for 48 h and weighed to the nearest 0.01 g. Mosses and lichens were collected from a 0.25 × 0.25 m area in the centre of each subplot used for collection of field-layer vegetation, sorted by species, dried and weighed. The biomass of each plant species (separately for woody and green tissues) was expressed in g d.w. m⁻², and these values were used to calculate site-specific C and N pools as described below.

2.5. Total C and N in plant tissues

Samples of 10 vascular plant species (listed in Table S2) were collected at each study site on 23–24 August 2012 from three haphazardly selected plant individuals of each species. Wood samples of top-canopy trees (including *Salix caprea* L.) were collected from twigs 12–18 mm in diameter, whereas all woody parts were collected from dwarf shrubs. Leaves/needles were separated from woody parts (only current-year foliage was collected from evergreen species) and the samples were dried (60 °C for 48 h) and milled for analysis of total C and N concentrations. The analyses were performed using high-temperature combustion (Vario MAX CN analyser, Elementar Analysensysteme GmbH, Germany) at the Department of Forest Sciences, University of Helsinki.

2.6. Calculation of C and N pools

Biomass data (by plant species and tissue fraction) were averaged for site-specific values and multiplied by respective species-, fraction- and site-specific concentrations of C and N (averaged from three samples per site). For fruits of dwarf shrubs, we used C and N concentrations in woody parts. For plant species that have not been analysed for C and N concentrations (5.8% of the biomass of top-canopy trees and understory and 22.6% of the biomass of field-layer vegetation), we used fraction- and site-specific C and N data from other species as outlined in Table S3. For mosses and lichens, we used 0.45% N and 44% C, based on data from Kevo, northern Finland (Manninen et al., 2013)

2.7. Data analysis

Main and interactive effects of plant species, tissue (foliage vs. woody parts) and study site on total C and N concentrations and C:N ratios were analysed using mixed model ANOVA. In this analysis, plant species and tissue were fixed factors, whereas the site and its interactions with species and tissue were random factors.

The relationships between the total C and N concentrations in plant tissues and pollutant load were explored using meta-analysis. Changes in C and N concentrations, as well as in their ratio, along the pollution gradient were quantified by calculating Pearson correlation coefficients with log-transformed concentrations of Cu in forest litter. To calculate effect sizes (ES), these coefficients were z-transformed and weighted by their sample size (i.e. by the number of study sites) using the standard procedure in the MetaWin programme (Rosenberg et al., 2000). In our study the negative ES value indicates that the parameter under study decreases with increase in pollution. We used a bootstrap estimate of the 95% confidence interval (CI₉₅), because the numbers of ES in individual groups were small (eight or ten). The effect was considered statistically significant if its CI₉₅ did not include zero. Meta-analysis was performed using random effects categorical models. The variation in the ESs between the classes of categorical variables was explored by calculating heterogeneity indices (Q_B), and testing them against χ^2 distribution.

The total aboveground plant biomass and the total C and N pools were regressed on Cu concentration in forest litter. The obtained equations were used to outline the areas where these response variables were affected by pollution from the smelter, and to estimate total losses of C and N pools from forests in northern Fennoscandia caused by pollution from non-ferrous industries. The upper limit for background Cu concentration in forest litter was considered to be 7 $\mu\text{g g}^{-1}$ (Tamminen et al., 2004); humus Cu concentrations were obtained from a recent biogeochemical survey (Reimann et al., 1998). Data from Forestry Plan for Murmansk Oblast (2008) and Finnish Statistical Yearbook of Forestry (Finnish Forest Research Institute, 2014) were used to calculate the average proportion of land area covered by forests (54%) in the Kola Peninsula and northern Finland. The ratio of aboveground to belowground plant biomass (78%:22%) was derived from the results on Canadian (Kurz and Apps, 1999), Russian (Shvidenko and Nilsson, 2002, 2003) and Finnish forests (Merilä et al., 2014).

3. Results

3.1. Concentrations of C and N in plant tissues

The total C and N concentrations, as well as C:N ratio, varied among species and between tissues, and this variation was differently expressed across study sites (Tables 2 and S2). The smallest site-specific C and N concentrations and C:N ratio in green tissues were measured in *Deschampsia flexuosa* shoots and were 42.0%, 1.05% and 19.6, respectively. The greatest C concentration (54.7%) and C:N ratio (48.3) were measured in *E. nigrum* leaves, and the greatest N concentration (2.35%) in *B. pubescens* leaves. In woody tissues, *Salix caprea* had the smallest site-specific C concentration (45.6%) and *E. nigrum* the greatest (54.8%). For N, the smallest value was measured in *Pinus sylvestris* (0.31%) and the largest in *V. vitis-idaea* (0.89%), whereas *V. vitis-idaea* had the smallest site-specific C:N ratio in its woody parts (54.9) and *P. sylvestris* twigs had the largest (161.7). Positive correlations between C and N concentrations within species were found in *E. nigrum* and

Table 2

Sources of variation in total C and N concentrations and C:N ratios in green and woody plant tissues in the Kola Peninsula (mixed model ANOVA, type III sums of squares).

Source	df	Significances of the effects (F/P)		
		Total C	Total N	C:N
Species	9	203.75/<0.001	8.17/<0.001	16.28/<0.001
Site	9	1.48/0.26	0.80/0.62	0.99/0.50
Tissue	1	20.07/0.002	459.95/<0.001	269.97/<0.001
Site × species	79	1.37/0.03	2.22/<0.001	1.22/0.11
Site × tissue	9	2.01/0.004	3.57/<0.001	5.95/<0.001
Error	405			

P. sylvestris woody tissues, while negative correlations were found in both the leaves and woody tissue of *V. myrtillus* (all $P < 0.05$, data not shown).

The total foliar C concentration, on average, increased with increasing pollution load, whereas total wood C concentration did not change (Fig. 2). The total foliar N decreased with increasing pollution load, although coniferous trees (*P. sylvestris* and *P. abies*), in contrast to other plants ($Q_B = 4.02$, $df = 1$, $P = 0.04$), demonstrated an increase in foliar N with increasing pollution load. The total N concentration in woody tissues increased with increasing pollution load. Consequently, the overall C:N ratio in the green parts of study plants generally increased with increasing pollution load, although gymnosperm and angiosperm species showed contrasting patterns ($Q_B = 6.67$, $df = 1$, $P = 0.01$). The C:N ratio in woody tissues decreased (Fig. 2). The average total C and N concentrations across study sites, plant species and tissues were 48.4% and 1.15%, respectively.

3.2. Aboveground biomass and C and N pools

The aboveground plant biomass, and, consequently, C and N pools, strongly decreased with an increase in the pollution load (Fig. 3). The smallest total aboveground biomass ($76\text{--}104\text{ g m}^{-2}$) was observed in industrial barrens, where coniferous species were practically absent. Dwarf shrub biomass was larger than that of coniferous or deciduous trees and shrubs at the site nearest to the smelter (1 N). The biomass of two other barren sites (6S and 8S) was almost entirely represented by mountain birches (Table S4). At the least polluted sites, the total aboveground biomass was $3091\text{--}4888\text{ g m}^{-2}$; conifers contributed 70–85%, followed by deciduous trees and shrubs (7–21%) and dwarf shrubs (5.5–6.3%). Mosses were found at all sites, whereas lichens were lacking or had a very minor biomass at sites within 10 km of the smelter (Table S4).

The site-specific C pool in aboveground phytomass ranged from $35\text{--}2333\text{ g m}^{-2}$ and that of N from $0.5\text{--}35.1\text{ g m}^{-2}$. At the site closest to the smelter, the low-stature trees and shrubs contributed 57.1% to the C pool. The corresponding values for field-layer vegetation, mosses and lichens were 40%, 2.9% and 0%, respectively. As the tree cover increased with the distance from the smelter, the proportion of the C pool in trees and shrubs reached 92–97%, while the contribution of field-layer vegetation decreased to $\leq 6.5\%$, and that of mosses and lichens decreased to $\leq 2.3\%$ and $\leq 1.5\%$, respectively. The proportion of the N pool in trees and shrubs ranged from 55–97% at the individual sites and the ranges for field-layer vegetation, mosses and lichens were 1.3–42%, 0.1–8.6% and 0–1.8%, respectively.

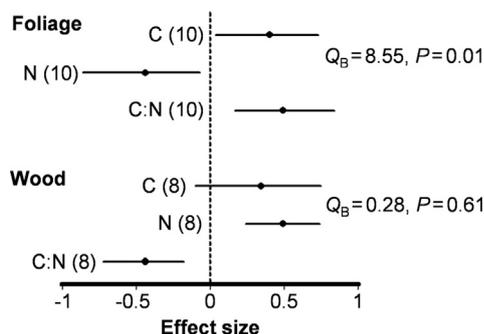


Fig. 2. Changes in the total C and N concentrations and C:N ratios in foliage and wood tissues of study plants in the pollution gradient: mean effect sizes (dots), 95% confidence intervals (horizontal lines) and sample sizes (in parentheses). A negative effect size indicates that the parameter under study decreases with pollution. The effect is significant if the 95% confidence interval does not overlap zero value. Q_B values indicate the difference between the changes in C and N concentrations.

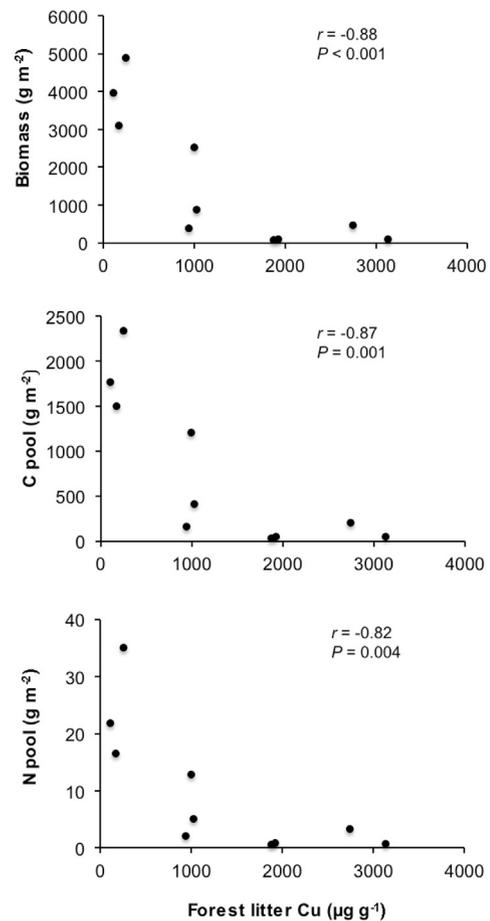


Fig. 3. Average site-specific values of living aboveground plant biomass, C pool and N pool in relationship to forest litter Cu concentration.

3.3. Losses of phytomass C stock caused by industrial air pollution

Based on the observed relationship between forest litter Cu concentration and the aboveground plant biomass (Fig. 3), we estimated a potential aboveground forest biomass of 8014 g m^{-2} in the area with the background litter Cu concentration ($7\text{ }\mu\text{g g}^{-1}$). Using a 78%:22% ratio for aboveground:belowground biomass, we estimated a potential total vegetation biomass of $10,274\text{ g m}^{-2}$ for these forests. Correspondingly, the potential total phytomass C pool in our study region, in the absence of pollution, is 4797 g m^{-2} . Given that the boreal forest zone in northern Finland, northern Norway and the Kola Peninsula had Cu concentrations in the humus layer that exceeded the background level of over an area of $107,200\text{ km}^2$ (Reimann et al., 1998), we estimated that industrial air pollution decreased the total plant biomass in this region to 89% of its potential value, and that the resulting loss of C stock was $4.24 \times 10^{13}\text{ g}$ (Table 3).

4. Discussion

4.1. Concentrations of C and N in plant tissues

The concentrations of C and N in plant tissues reflect CO_2 and N assimilation that determines plant growth and biomass production. These processes and fluxes of N and C assimilates depend on a supply of resources from, and conditions in, the environment (Lawlor, 2002). Therefore, pollution-induced changes in the abiotic environment are likely to have both direct and indirect effects on the average C and N concentrations and their ratios in plant tissues. At our study sites, C

Table 3

Losses of plant biomass and phytomass C stock in boreal forest zone of northern Fennoscandia caused by industrial air pollution.

Cu in humus layer, $\mu\text{g g}^{-1}$		Area ^a , km^2	Estimated ^b alive phytomass in forested areas, g m^{-2}		Estimated ^d loss of alive phytomass, g	Estimated ^e loss of C, g
Min–max	Mean		Aboveground	Total ^c		
4–7	5.5	31,500	8340	10,690	–	–
7–9	8.0	30,600	7840	10,050	3.7×10^{12}	1.8×10^{12}
9–14	11.5	38,400	7350	9420	17.7×10^{12}	8.6×10^{12}
14–28	21	22,300	6550	8400	22.5×10^{12}	10.9×10^{12}
28–110	69	12,100	4960	6360	25.5×10^{12}	12.3×10^{12}
110–2460	1285	3800	1050	1350	18.3×10^{12}	8.8×10^{12}
Total		138,700 ^f	–	–	87.7×10^{12}	42.4×10^{12}

^a After Reimann et al. (1998).^b Calculated from the regression equation: Aboveground biomass (g d.w. m^{-2}) = $10,616 - 1337 \times \ln(\text{concentrations of Cu in humus layer, } \mu\text{g g}^{-1})$.^c Calculated as aboveground biomass divided by 0.78, i.e. by average proportion of the aboveground biomass in boreal forests (after Kurz and Apps, 1999; Shvidenko and Nilsson, 2002, 2003; Merilä et al., 2014).^d Calculated as the difference between potential total biomass in absence of contamination ($10,274 \text{ g m}^{-2}$; for explanations, see text) and the estimated total biomass, corrected for the average proportion of land area covered by forests (54%, after Forestry Plan for Murmansk Oblast, 2008 and Finnish Forest Research Institute, 2014).^e Calculated as the estimated loss of biomass multiplied by the average total C concentration across plant species and tissues (48.4%).^f Total affected area (i.e. the area where concentrations of Cu in humus layer exceeded $7 \mu\text{g g}^{-1}$) is $107,200 \text{ km}^2$.

and N concentrations in green tissues were decoupled, suggesting that different factors may be regulating C and N assimilation and translocation processes under pollution stress.

The soils in the vicinity of the Monchegorsk smelter (especially the organic surface layer) have become N-depleted (Lukina and Nikonov, 1999; Rautio and Huttunen, 2003). This depletion is attributed to the reduction in biomass production and subsequently soil organic matter (Lozano and Morrison, 1981; Rautio and Huttunen, 2003), enhanced erosion of surface soil (Kozlov et al., 2009), impaired microbial activity and changes in the structure of the soil microbial community (Evdokimova, 2000; Mälkönen et al., 1999) due to impacts of SO_2 and heavy metals. Evdokimova (2000) showed that N_2 -fixing, nitrifying, cellulolytic and cyanobacteria were totally lost within 7–10 km from the smelter, which explained the pollution-related disturbance of plant litter decomposition (Kozlov and Zvereva, 2015), N mineralization and biological N_2 fixation in the ecosystem. This pollution-induced N depletion explains the overall decrease in foliar N concentrations with increasing pollution, although direct toxic effects of heavy metals to rhizomes, roots and mycorrhiza (Mälkönen et al., 1999) may also have contributed to this effect.

In contrast to other species, conifers demonstrate an increase in foliar N with increase in pollution. This may be explained by stomatal uptake of NO_x by coniferous trees (Ammann et al., 1999; Manninen and Huttunen, 2000) and by the reduction in the number of needle age-classes in polluted areas, with the subsequent allocation of nutrients into remaining needles (Rautio et al., 1998a; Stjernquist et al., 1998). Note that forest litter Cu concentration, which was used an overall indicator of pollution load, correlated well with ambient SO_2 concentration in the area (Kozlov et al., 2009). Increases in needle total S concentration towards the smelter (Rautio and Huttunen, 2003) emphasize the impact of gaseous pollutants on element concentrations in the evergreen needles as well as on foliar health in terms of both visible and invisible damage.

We measured N concentrations in branches of trees, which may have 5–10 times the N concentration found in stem wood (Merilä et al., 2014; Wirth et al., 2002). Therefore, our results overestimate the actual N pools, and this overestimation increases with increase in the proportion of trees in site-specific phytomass. Still, our results show significant effects of industrial pollution on ecosystem N dynamics, given that the aboveground N pool at the most polluted sites only amounted to 5.1 g N m^{-2} at its largest, while the average total aboveground phytomass N stock in coniferous forests of northern Finland is 25 g N m^{-2} (Merilä et al., 2014).

The overall increase in C concentration in green tissues with increasing pollution load may partly be explained by enhanced photosynthesis with decreasing shading. Nevertheless, visible and microscopic needle

injuries, reduced number of needle age-classes (Rautio et al., 1998b; Rautio and Huttunen, 2003), reduced radial growth of *P. sylvestris* (Nöjd and Reams, 1996) and the reduced aboveground plant biomass in the surroundings of the smelter indicate overall negative effects of pollution on net primary production. The opposite changes in C:N ratio found in green and woody tissues further hint that pollution affects the translocation of C and N within a plant. For example, non-structural carbohydrates may accumulate in the foliage of plants growing near the smelter due to reduced soil N supply and subsequent decreases in the amount and activity of the N-containing Rubisco enzyme in leaves (DeLucia et al., 1985; Tissue et al., 1993). The overall increase in wood N concentration with increasing pollution load and the positive correlations between wood C and N concentrations in the woody tissues of evergreen *E. nigrum* and *P. sylvestris* may suggest enhanced cortical photosynthesis under pollution stress (Manninen et al., 2009; Wittman et al., 2007). This enhancement would also be facilitated by increased light availability for the persisting plants due to the disappearance of the top-canopy trees.

Still, the detected changes in concentrations of C and N in plant tissues along the pollution gradient, although physiologically important, were minor when compared to the drastic reduction in aboveground plant biomass with increasing pollution load. Moreover, the plants from our study sites cannot be considered as N deficient, because their foliar total N concentrations were in the same range as those from plants in unpolluted forests (Hansen et al., 2006; Laine and Henttonen, 1987; Manninen et al., 1997a; Manninen and Huttunen, 2000; Sveinbjörnsson et al., 1992). This is because the higher light availability and lower nutrient availability in highly polluted compared to less polluted sites may have promoted greater allocation of C below ground to support root growth and nutrient uptake.

4.2. Biomass reductions

Elevated S concentration in *P. sylvestris* needles (Manninen et al., 1997a, 1997b) and elevated Cu concentration in the humus layer (Reimann et al., 1998) have been found at distances of up to 200 km from the Monchegorsk smelter. However, visible and microscopic symptoms of multiple stresses (S, Cu, Ni, acid deposition, O_3 and frost) in needles of *P. sylvestris* and reduced numbers of needle age-classes were clearly detectable only within 40 km from the smelter (Rautio et al., 1998b; Rautio and Huttunen, 2003), and reduced radial growth of *P. sylvestris* was only recorded within 30 km southwest of the smelter (Nöjd and Reams, 1996). Therefore, at the planning stage of the study, we did not expect that relatively low levels of industrial pollution, occurring beyond 30–40 km from the smelter and causing no visible injuries and only slight physiological disturbance, would have a drastic

effect on plant biomass. Another surprising feature was that the greatest total aboveground biomass found in our study area was as low as the smallest aboveground plant biomass in the similarly aged semi-natural forests under conventional management in unpolluted regions of northern Finland (Merilä et al., 2014). Keeping in mind the similar environmental conditions in northern Finland and in the Kola Peninsula, we suggest that this difference resulted primarily from long-term impacts of industrial emissions on forests in the central part of the Kola Peninsula. Thus, although an overall decrease in productivity in the vicinities of industrial sources of atmospheric pollutants is relatively well documented (Zvereva and Kozlov, 2012), our study is the first to demonstrate the extent and severity of the adverse effects of industrial pollution on the total biomass of forest plants at the regional scale. In our most polluted sites, the total aboveground plant biomass was reduced to 1.5–3.5% of that observed in the least polluted sites, and comprised about 1% of the potential aboveground plant biomass.

Our estimate of the potential aboveground plant biomass in forests of our study region (8014 g m^{-2}) fits well the results of measurements conducted in unpolluted forests. In northern Finland, the total aboveground biomass of vegetation in coniferous forests varied from $4960\text{--}12,280 \text{ g m}^{-2}$ (Havas and Kubin, 1983; Merilä et al., 2014). In slightly polluted forests of the Kola Peninsula, total aboveground biomass in coniferous forests aged 100+ years varied from $4100\text{--}12,800 \text{ g m}^{-2}$ (Manakov and Nikonov, 1979; Zybchenko and Ivanchikov, 1978). Shvidenko and Nilsson (2002, 2003) estimated an average aboveground phytomass 4860 g m^{-2} for forest tundra and northern and sparse taiga, and 9140 g m^{-2} for middle taiga. Hence, overestimation of the average potential biomass is unlikely in the studied forest ecosystems; as is overestimation of pollution-induced losses in plant biomass at the regional scale.

Pollution can reduce plant growth, but it can also modify biomass allocation to aboveground and belowground plant parts. A decrease in the root–shoot ratio is a general response of herbaceous plants to pollutants; however, no quantitative data on the root–shoot ratio exist for mature trees (Zvereva et al., 2010). Birch seedlings showed higher biomass allocation to roots when growing in polluted relative to unpolluted soils (Bojarczuk et al., 2002), while an application of acid rain and heavy metals to birch seedlings during two months caused no statistically significant changes in the root–shoot ratio (Koricheva et al., 1997). Thus, the use of the 78%:22% ratio for aboveground:belowground biomass in our study region, as estimated for unpolluted boreal forests (Kurz and Apps, 1999; Merilä et al., 2014; Shvidenko and Nilsson, 2002, 2003), was unlikely to introduce strong bias in our calculations. It is noted, however, that in the northernmost forests understory roots and rhizomes may account for up to 50% of the stand total fine root biomass (Helmisaari et al., 2007). This emphasizes the need to measure belowground biomass given the differences in biomass of each plant group in relation to pollution load and natural site-specific environmental factors.

4.3. Carbon losses from forests due to pollution

Fires and insect pests impose the major disturbances in northern forests (Kurz and Apps, 1999; Shvidenko and Nilsson, 2003). However, the land area affected by pollution in the boreal forest zone of Fennoscandia, due to nearly 80 years of industrial activity in the Kola Peninsula ($107,200 \text{ km}^2$, as estimated from Cu concentrations in forest litter: Reimann et al., 1998), exceeded by a factor of 500 the total area of extensive forest fires in the north-western Russia in a normal year in early 2000s (which averaged ca. 200 km^2 : Selikhovkin, 2005). The direct and indirect losses of phytomass C from outbreaks of insect pests in forests of north-western Russia between 1953 and 1998 were, on average, 0.013 year^{-1} (Selikhovkin, 2009). Assuming that fires destroy aboveground plant biomass completely, while pollution reduces it by 11%, on average, we conclude that the direct losses of the C pool at the regional scale due to industrial pollution from the smelters located in

the Kola peninsula, combined with the losses in C sequestration capacity due to pollution-induced decline in plant biomass, are about the same magnitude as the cumulative losses to forest fires and insect pests over the past 50 years. However, the effects of industrial pollution on C pool are quite likely several times larger than the effects of fires and pests, in particular because the estimated loss of $4.24 \times 10^{13} \text{ g C}$ did not account for C loss due to extensive soil erosion in the polluted areas, with the soil humus layer being reduced from 30–35 mm in unpolluted forests to 0–10 mm in industrial barrens (Kozlov et al., 2009).

The Monchegorsk smelter is not the sole point polluter in the boreal forest zone. While the emissions from Canadian smelters (Trail, Sudbury, Wawa) have decreased markedly since 1970s, several large point-sources, in particular the Norilsk smelter in northern Siberia (emitting 2 000 000 t of SO_2 , 9 000 t NO_x , 600 t of Cu and 500 t of Ni annually), continue to contaminate the boreal zone (Kozlov et al., 2009, and references therein). Given this, the overall impact of the industrial polluters on ecosystem C and N pools should be taken into account when assessing the C and N dynamics in northern boreal forests because of the marked long-term negative effects of their emissions on the structure and productivity of plant communities.

At the global scale, 8% of the forested areas of the World received $>1 \text{ kg H}^+ \text{ ha}^{-1} \text{ year}^{-1}$ as S in 1985, and current estimates are that 17% of the world's forested areas will receive this pollution load by 2050. Similarly, 24% of the global forest was exposed to O_3 concentrations exceeding 60 ppb by 1990, and this proportion is expected to increase to 50% of the global forest by 2100 (Fowler et al., 1999). These changes will have global consequences for primary productivity, C and N cycles and other characteristics of forest ecosystems. This is emphasized by the fact that about half of the losses in the aboveground C and N pools in the study area occurred in slightly contaminated areas outside the zone of visible forest damage.

5. Conclusions

We demonstrated that the long-term impacts of atmospheric emissions from a large industrial point source, the Ni–Cu smelter in Monchegorsk, resulted in extensive losses of phytomass C stock and of C sequestration capacity from the affected forests. At the regional scale, the consequences of industrial pollution are about the same magnitude (but presumably larger) than the cumulative effects of fires and outbreaks of insect pests over the past 50 years. Despite a recent reduction in emissions, no natural recovery has yet been observed in the most polluted sites due to legacy effects such as extremely high concentrations of metals and shortage of nutrients in the thin, eroded soil (Zverev, 2009). The complete leaching of metals accumulated in the upper soil horizons near Monchegorsk will take 160–270 years for Ni and 100–200 years for Cu, after the cessation of emissions (Barcan, 2002); thus, the consequences of past industrial activities will continue to affect ecosystem processes well into the future. Therefore, air pollution effects on terrestrial ecosystems should be taken into account in the analysis and modelling of regional and global C and N budgets.

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.scitotenv.2015.07.097>.

Acknowledgements

The study was financially supported by the Academy of Finland (project 124152) and by the University of Helsinki. We thank P. Kauppi, A. Legge, E. Vorobeichik and E. Zvereva for their useful comments to an earlier version of the manuscript.

References

- Ammann, M., Siegwolf, R., Pichlmayer, F., Suter, M., Saurer, M., Brunold, C., 1999. Estimating the uptake of traffic-derived NO_2 from ^{15}N abundance in Norway spruce needles. *Oecologia* 118, 124–131.

- Barcan, V.S., 2002. Leaching of nickel and copper from soil contaminated by metallurgical dust. *Environ. Int.* 28, 63–68.
- Bojarczuk, K., Karolewski, P., Oleksyn, J., Kieliszewska-Rokicka, B., Żytkowiak, R., Tjoelker, M.G., 2002. Effect of polluted soil and fertilisation on growth and physiology of silver birch (*Betula pendula* Roth.) seedlings. *Pol. J. Environ. Stud.* 11, 483–492.
- Bytnerowicz, A., Arbaugh, M., Schilling, S., Frączek, W., Alexander, D., 2008. Ozone distribution and phytotoxic potential in mixed conifer forests of the San Bernardino Mountains, southern California. *Environ. Pollut.* 155, 398–408.
- de Vries, W., Dobberty, M.H., Solberg, S., van Dobben, H.F., Schaub, M., 2014. Impacts of acid deposition, ozone exposure and weather conditions on forest ecosystems in Europe: an overview. *Plant Soil* 380, 1–45.
- DeLucia, E.H., Sasek, T.W., Strain, B.R., 1985. Photosynthetic inhibition after long-term exposure to elevated levels of atmospheric carbon dioxide. *Photosynth Res.* 7, 175–184.
- Eeva, T., Belskii, E., Gilyazov, A.S., Kozlov, M.V., 2012. Pollution impacts on bird population density and species diversity at four non-ferrous smelter sites. *Biol. Conserv.* 150, 33–42.
- Evdokimova, G.A., 2000. The impact of air pollution on the northern taiga forests of the Kola Peninsula, Russian Federation. In: Innes, J.L., Oleksyn, J. (Eds.), *Forest dynamics in heavily polluted regions*. CABI Publishing, Wallingford, UK, pp. 67–76.
- Felzer, B., Kicklighter, D., Mellilo, J., Wang, C., Zhuang, Q., Prinn, R., 2004. Effects of ozone on net primary production and carbon sequestration in the conterminous United States using a biogeochemistry model. *Tellus* 56B, 230–248.
- Finnish Forest Research Institute, 2014. Finnish Statistical Yearbook of Forestry (In Finnish with English summary). <http://www.metla.fi/julkaisut/metsatilastollinen/vsk/tilastovsk-sisalto.htm> (accessed 13 March 2015).
- Fischer, T., Bergmann, C., Hüttl, R.F., 1995. Soil carbon and nitrogen budget in Scots pine (*Pinus sylvestris* L.) stands along an air pollution gradient in eastern Germany. *Water Air Soil Pollut.* 85, 1671–1676.
- Forestry Plan for Murmansk Oblast, 2008. Vol. 1: explanatory notes. Rosgiproles, Moscow (in Russian available at: <http://mpr.gov-murman.ru/documents/lesplan/>; accessed on 8 March 2015).
- Fowler, D., Cape, J.N., Coyle, M., Flechard, C., Kuylentierna, D., Hicks, K., Derwent, D., Johnson, C., Stevenson, D., 1999. The global exposure of forests to air pollutants. *Water Air Soil Pollut.* 116, 5–32.
- Freedman, B., 1989. *Environmental ecology*. Academic Press, New York.
- Goodale, C.L., Apps, M.J., Birdsey, R.A., Field, C.B., Heath, L.S., Houghton, R.A., et al., 2002. Forest carbon sinks in the northern hemisphere. *Ecol. Appl.* 12, 891–899.
- Graven, H.D., Keeling, R.F., Piper, S.C., Patra, P.K., Stephens, B.B., Wofsy, S.C., et al., 2013. Enhanced seasonal exchange of CO₂ by northern ecosystems since 1960. *Science* 341, 1085–1089.
- Hansen, A.H., Jonasson, S., Michelsen, A., Julkunen-Tiitto, R., 2006. Long-term experimental warming, shading and nutrient addition affect the concentration of phenolic compounds in arctic-alpine deciduous and evergreen dwarf shrubs. *Oecologia* 147, 1–11.
- Havas, P., Kubin, E., 1983. Structure, growth and organic matter content in the vegetation cover of an old spruce forest in Northern Finland. *Ann. Bot. Fenn.* 20, 115–149.
- Helmisääri, H.-S., Derome, J., Nöjd, P., Kukkola, M., 2007. Fine root biomass in relation to site and stand characteristics in Norway spruce and Scots pine stands. *Tree Physiol.* 27, 1493–1504.
- Kauppi, P., Mielikäinen, K., Kuusela, K., 1992. Biomass and carbon budget of European forests, 1971–1990. *Science* 256, 70–74.
- Koricheva, J., Roy, S., Vranjic, J.A., Haukioja, E., Hughes, P.R., Hänninen, O., 1997. Antioxidant responses to simulated acid rain and heavy metal deposition in birch seedlings. *Environ. Pollut.* 95, 249–258.
- Kozlov, M.V., Zvereva, E.L., 2011. A second life for old data: global patterns in pollution ecology revealed from published observational studies. *Environ. Pollut.* 159, 1067–1075.
- Kozlov, M.V., Zvereva, E.L., 2015. Decomposition of birch leaves in heavily polluted industrial barrens: relative importance of leaf quality and site of exposure. *Environ. Sci. Pollut. Res.* <http://dx.doi.org/10.1007/s11356-015-4165-8>.
- Kozlov, M.V., Zvereva, E.L., Zverev, V., 2009. Impacts of point polluters on terrestrial biota. *Environmental Pollution* 15. Springer, Dordrecht. <http://dx.doi.org/10.1007/978-90-481-2467-1>.
- Kurz, W.A., Apps, M.J., 1999. A 70-year retrospective analysis of carbon fluxes in the Canadian forest sector. *Ecol. Appl.* 9, 526–547.
- Laine, K.M., Henttonen, H., 1987. Phenolics/nitrogen ratios in the blueberry *Vaccinium myrtillus* in relation to temperature and microtine density in Finnish Lapland. *Oikos* 50, 389–395.
- Lawlor, D.W., 2002. Carbon and nitrogen assimilation in relation to yield: mechanisms are the key to understanding production systems. *J. Exp. Bot.* 53, 773–787.
- Le Quéré, C., Andres, R.J., Boden, T., Conway, T., Houghton, R.A., House, J.I., et al., 2013. The global carbon budget 1959–2011. *Earth Syst. Sci. Data* 5, 165–185. <http://dx.doi.org/10.5194/essd-5-165-2013>.
- Lorenz, K., Lal, R., 2010. Carbon sequestration in forest ecosystems. Springer, Dordrecht http://dx.doi.org/10.1007/978-90-481-3266-9_1.
- Lozano, F.C., Morrison, I.K., 1981. Distribution of hardwood nutrition by sulphur dioxide, nickel and copper air pollution near Sudbury, Canada. *J. Environ. Qual.* 10, 198–204.
- Lukina, N.V., Nikonov, V.V., 1999. Pollution-induced changes in soils subjected to intense air pollution. In: Nikonov, V.V., Koptsik, G.N. (Eds.), *Acidic deposition and forest soils*. Kola Science Centre, Apatity, pp. 79–126 (in Russian).
- Magnani, F., Mencuccini, M., Borghetti, M., Berbigie, P., Berninger, F., Delzon, S., et al., 2007. The human footprint in the carbon cycle of temperate and boreal forests. *Nature* 447, 848–850.
- Mälkönen, E., Derome, J., Fritze, H., Helmisääri, H.S., Kukkola, M., Kytö, M., et al., 1999. Compensatory fertilization of Scots pine stands polluted by heavy metals. *Nutr. Cycl. Agroecosyst.* 55, 239–268.
- Manakov, K.N., Nikonov, V.V., 1979. Primary biological productivity of spruce forests in the Kola Peninsula. *Bot. Zh.* 64, 232–241 (in Russian).
- Manninen, S., Huttunen, S., 2000. Response of needle sulphur and nitrogen concentrations of Scots pine versus Norway spruce to SO₂ and NO₂. *Environ. Pollut.* 107, 421–436.
- Manninen, S., Huttunen, S., Perämäki, P., 1997a. Needle S fractions and S to N ratios as indices of SO₂ deposition. *Water Air Soil Pollut.* 95, 277–298.
- Manninen, S., Huttunen, S., Kontio, M., 1997b. Accumulation of sulphur in Scots pine needles in the subarctic. *Water Air Soil Pollut.* 95, 147–164.
- Manninen, S., Huttunen, S., Vanhatalo, M., Pakonen, T., Hämäläinen, A., 2009. Inter- and intra-specific responses to elevated ozone and chamber climate in northern birches. *Environ. Pollut.* 157, 1679–1688.
- Manninen, S., Sassi, M.K., Lovén, K., 2013. Effects of nitrogen oxides on ground vegetation, *Pleurozium schreberi* and the soil beneath it in urban forests. *Ecol. Indic.* 24, 485–493.
- Matyssek, R., Wieser, G., Calafapietra, C., de Vries, W., Dizengremel, P., Ernst, D., et al., 2012. Forests under climate change and air pollution: gaps in understanding and future directions for research. *Environ. Pollut.* 160, 57–65.
- Merilä, P., Mustajärvi, K., Helmisääri, H.S., Hilli, S., Lindroos, A.J., Nieminen, T.M., et al., 2014. Above- and below-ground N stocks in coniferous boreal forests in Finland: Implications for sustainability of more intensive biomass utilization. *For. Ecol. Manag.* 311, 17–28.
- Nöjd, P., Reams, G.A., 1996. Growth of Scots pine across pollution gradient on the Kola Peninsula, Russia. *Environ. Pollut.* 93, 313–325.
- Pan, Y., Birdsey, R.A., Fang, J., Houghton, R., Kauppi, P.E., Kurz, W.A., et al., 2011. A large and persistent carbon sink in the World's forests. *Science* 333, 988–993.
- Prank, M., Sofiev, M., Denier van der Gon, H.A.C., Kaasik, M., Ruuskanen, T.M., Kukkonen, J., 2010. A refinement of the emission data for Kola Peninsula based on inverse dispersion modelling. *Atmos. Chem. Phys.* 10, 10849–10865.
- Rautio, P., Huttunen, S., 2003. Total vs. internal foliar element concentrations in Scots pine along a sulphur and metal pollution gradient. *Environ. Pollut.* 122, 273–289.
- Rautio, P., Huttunen, S., Lamppu, J., 1998a. Seasonal foliar chemistry of northern Scots pines under sulphur and heavy metal pollution. *Chemosphere* 37, 271–287.
- Rautio, P., Huttunen, S., Kukkola, E., Peura, R., Lamppu, J., 1998b. Deposited particles, element concentrations, and needle injuries on Scots pine along an industrial pollution transect in northern Europe. *Environ. Pollut.* 103, 81–89.
- Reimann, C., Åyräs, M., Chekushin, V., Bogatyrev, I., Boyd, R., de Caritat, P., et al., 1998. Environmental geochemical atlas of the central Barents region. Geological Survey of Norway, Trondheim.
- Rigina, O., Kozlov, M.V., 2000. Pollution impact on sub-Arctic northern taiga forests in the Kola peninsula, Russia. In: Innes, J.L., Oleksyn, J. (Eds.), *Forest dynamics in heavily polluted regions*. CABI Publishing, Wallingford, UK, pp. 37–65.
- Rosenberg, M.S., Adams, D.C., Gurevitch, J., 2000. MetaWin: statistical software for meta-analysis. Version 2.0. Sinauer, Sunderland.
- Schütt, P., Cowling, E.B., 1985. Waldsterben – a general decline of forests in central Europe: symptoms, development, and possible causes of a beginning breakdown of forest ecosystems. *Plant Dis.* 69, 548–558.
- Selikhovkin, A.V., 2005. Main disturbance factors in north-west Russian forests: structure and databases. *Scand. J. For. Res.* 20, 27–32.
- Selikhovkin, A.V., 2009. Can outbreaks of dendrophagous insects make a considerable impact on the biosphere? *Biosphere* 1, 72–81 (In Russian, English summary).
- Shvidenko, A., Nilsson, S., 2002. Dynamics of Russian forests and the carbon budget in 1961–1998: an assessment based on long-term forest inventory data. *Clim. Chang.* 55, 5–37.
- Shvidenko, A., Nilsson, S., 2003. A synthesis of the impact of Russian forests on the global carbon budget for 1961–1998. *Tellus* 55B, 391–415.
- Stern, D.I., 2006. Reversal of the trend in global anthropogenic sulfur emissions. *Glob. Environ. Chang. Hum. Policy Dimens.* 16, 207–220.
- Stjernquist, I., Nihlgård, B., Filiptchouk, A.N., Strakhov, V.V., 1998. Soil and forest vitality as affected by air pollutants on the Kola Peninsula. *Chemosphere* 36, 1119–1124.
- Sveinbjörnsson, B., Nordell, O., Kauhanen, H., 1992. Nutrient relations of mountain birch growth at and below the elevational tree-line in Swedish Lapland. *Funct. Ecol.* 6, 213–220.
- Tamminen, P., Starr, M., Kubin, E., 2004. Element concentrations in boreal, coniferous forest humus layers in relation to moss chemistry and soil factors. *Plant Soil* 259, 51–58.
- Tissue, D.T., Thomas, R.B., Strain, B.R., 1993. Long-term effects of elevated CO₂ and nutrients on photosynthesis and rubisco in loblolly pine seedlings. *Plant Cell Environ.* 16, 859–865.
- Treshow, M., 1984. *Air pollution and plant life*. Wiley, New York.
- Tuovinen, J.P., Laurila, T., Lähtilä, H., 1993. Impact of the sulphur dioxide sources in the Kola Peninsula on air quality in northernmost Europe. *Atmos. Environ.* 27, 1379–1395.
- Vancura, K., Raben, G., Gorzelak, A., Mikulowski, M., Caboun, V., Oleksyn, J., 2000. Impact of air pollution on the forests of Central and Eastern Europe. In: Innes, J.L., Oleksyn, J. (Eds.), *Forest dynamics in heavily polluted regions*. CABI Publishing, Wallingford, UK, pp. 121–146.
- Vorobeichik, E.L., Kozlov, M.V., 2012. Impact of point polluters on terrestrial ecosystems: methodology of research, experimental design, and typical errors. *Russ. J. Ecol.* 43, 89–96.
- Wirth, C., Schulze, E.D., Lühker, B., Grigoriev, S., Siry, M., Hardes, G., et al., 2002. Fire and site type effects on the long-term carbon and nitrogen balance in pristine Siberian Scots pine forests. *Plant Soil* 242, 41–63.
- Wittman, C., Matyssek, R., Pfanz, H., Humar, M., 2007. Effects of ozone impact on the gas exchange and chlorophyll fluorescence of juvenile birch stems (*Betula pendula* Roth.). *Environ. Pollut.* 150, 258–266.
- Zverev, V.E., 2009. Mortality and recruitment of mountain birch (*Betula pubescens* ssp. *czerepanovii*) in the impact zone of a copper-nickel smelter in the period of

- significant reduction of emissions: the results of 15-year monitoring. *Russ. J. Ecol.* 40, 254–260.
- Zvereva, E.L., Kozlov, M.V., 2012. Changes in the abundance of vascular plants under the impact of industrial air pollution: a meta-analysis. *Water Air Soil Pollut.* 223, 2589–2599.
- Zvereva, E.L., Roitto, M., Kozlov, M.V., 2010. Growth and reproduction of vascular plants under pollution impact: a synthesis of existing knowledge. *Environ. Rev.* 18, 355–367.
- Zyabchenko, S.S., Ivanchikov, A.A., 1978. Zonal specificity of formation of Scots pine forests with bilberry in Karelia and Kola Peninsula and the dynamics of their biomass structure. In: Kazimirov, N.I., Zyabchenko, S.S. (Eds.), *Formation and productivity of Scots pine forests of Karelian Republic and Murmansk oblast*. Forest Institute, Petrozavodsk, pp. 30–75 (in Russian).