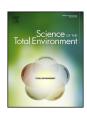
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# Consequences of long-term severe industrial pollution for aboveground carbon and nitrogen pools in northern taiga forests at local and regional scales



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#### 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 km<sup>2</sup>.
- The regional loss of phytomass carbon stock was estimated at  $4.24 \times 10^{13}$  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.

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#### 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 C and C 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 C u concentration in forest litter) caused C to increase in foliage but C remained unchanged in wood, while C demonstrating strong effects of pollution on resource translocation between green and woody tissues. The aboveground C and C pools were primarily governed by plant biomass, which strongly decreased with an increase in pollution load. In our study sites (located C 1.6–39.7 km from the smelter) living aboveground plant biomass was C to C 4888 g m $^{-2}$ , and C and C pools ranged C 35–2333 g C m $^{-2}$  and C 35.1 g C 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 C 107,200 km $^{-2}$ , and the total (aboveground and belowground) loss of phytomass C stock amounts to C 4.24 × 10 $^{13}$  g C 0.04 results emphasize the need to account for the overall impact of industrial polluters on ecosystem C and C pools when

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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.

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#### 1. Introduction

Annual global carbon dioxide (CO<sub>2</sub>) emissions from fossil fuel combustion and industry reached 9.5  $\pm$  0.5  $\times$  10<sup>15</sup> 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  $1.14 \times 10^7$  km<sup>2</sup> and are an important C storage globally. They provided a C sink of  $5.0 \pm 1.0 \times 10^{14}$  g C year<sup>-1</sup> 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 (SO<sub>2</sub>) 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 (O<sub>3</sub>), formed from the emissions of nitrogen oxides (NO<sub>x</sub>), 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 O<sub>3</sub> 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),  $SO_2$  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 aircleaning facilities until 1968. The annual emissions of  $SO_2$  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  $SO_2$  concentrations exceeded 60  $\mu g$  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

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