

Impact of alkaline cement-dust pollution on boreal *Pinus sylvestris* forest communities: a study at the bryophyte synusia level

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The elementary structural units where changes of plant communities come about are synusia and studies of successional processes induced by external factors should focus on that scale. Synusia is a structural part of a plant community inhabiting a special microhabitat, with a specific floristic composition and consisting of species that belong to the same stratum and that do not differ fundamentally in either periodicity or way of exploitation of their environment. We studied the responses of the moss synusia to the cement kiln dust pollution in *Myrtillus* site-type forests in the vicinity of the Kunda cement plant (North Estonia). The synusia were clustered into eight societies. The species content of the bryophyte synusia changed completely along the alkaline pollution gradient. Synusia of *Dicranum polysetum*–*Pleurozium schreberi*, *Aulacomnium palustre*–*Hylocomium splendens* and *Ptilium crista-castrensis*–*Hylocomium splendens* societies, characteristic of unpolluted forests, were replaced gradually by the synusia of *Rhytidiadelphus squarrosus*–*Hylocomium splendens*, *Climacium dendroides*–*Hylocomium splendens* and *Rhodobryum roseum*–*Rhytidiadelphus triquetrus* societies as pollution increased. *Brachythecium rutabulum*–*Rhytidiadelphus triquetrus* and *Fissidens dubius*–*Rhytidiadelphus triquetrus* societies were indicative of forests with heavy alkaline pollution. The composition of epiphytic bryophytes also changed almost completely along the pollution gradient; *Fissidens adianthoides*, *Tortula ruralis* and *Barbula unguiculata* were indicative of the heavily-polluted zone. We conclude that alkaline pollution has a clearly detectable impact on the forests' bryophyte synusia in terms of both species composition and typological structure.

Introduction

The vegetation damage caused by air pollution is a serious problem in many industrialized countries (e.g. Taylor & Johnson 1994, McLaughlin & Percy 1999, Bell & Treshow 2002,

Cheren'kova 2003). Acidic air pollution has been the most common stressor for plants (e.g. Falkengren-Grerup 1986, Smith 1990, Driscoll *et al.* 2001), while alkaline emission dominates in some regions and has also a remarkable impact on the plant community structure (Haapala *et al.*

1996a, 1996b, Kubíková 1981, Lameed & Ayo-dele 2008, Sujetoviené 2008).

Major sources of alkaline pollution are limestone quarries and cement plants (Teras 1984, Farmer 1993, Haapala *et al.* 1996a, 1996b, 2001, Zerrouqi *et al.* 2008). The effect intensity depends on quantity of deposited particles, chemical composition of the pollutant, distance from emitters, dominant wind direction, orography of the area, frequency and duration of emission (Farmer 2002).

In our previous paper dealing with the impact of alkaline pollution on the *Myrtillus* site-type Scots-pine boreal forests (Paal *et al.* 2013), we showed that the forest litter pH in the polluted zone around the Kunda cement factory reached 7.1–7.4 and the Ca content was ten-fold higher than in unpolluted areas. We detected a linear increase in species richness from habitats on acidic soils (unpolluted) towards those on alkalinized (polluted) soils. The initially typical boreal forest communities have notably nemoralized, i.e. they include beside numerous calcicolous species not typical of these communities in the natural state also several warmth- and nutrient-demanding species inherent mainly to broad-leaved forests of central and southern Europe. At the same time, the establishment of several endangered calcicolous species shows their dispersal potential but narrow soil pH niche.

Analyzing the pollution impact on multilayer forest communities, we should consider that the vegetation layers react somewhat independently to anthropogenic factors. This follows from the basic nature of the layers, consisting of species with different growth forms and ecological tolerance. The relative independence of responses of the moss layer from those of the taller phanerogam layers has been documented in numerous papers (e.g. During & Verschuren 1988, Paal 1995, Ewald 2000). But layers of natural plant communities are usually not homogeneous; if the considered layer includes various species, it represents a mosaic of predictable combinations of species termed *synusiae*. Barkman (1973: 452) defined *synusia* as a structural part of a plant community “inhabiting (a) a special microhabitat, with (b) a specific floristic composition and consisting of species that (c) belong to the same stratum and that do not differ fundamentally

in either (d) periodicity or (e) way of exploitation of their environment”. Vegetation dynamics is expressed simultaneously by two processes: changes of species content and changes in communities structure (Norin 1979). Because the elementary structural units where changes of communities come about are *synusiae* (Du Rietz 1965, Komarova 1993, Decocq 2002), studies to document the successional processes induced by external factors should focus on that scale (Lippmaa 1933, Norin 1979, Werffeli *et al.* 1997, Decocq 2002).

Bryophytes and epiphytic lichens are well known and widely used indicators of air pollution (Addison & Puckett 1980, Carlberg *et al.* 1983, Nimis *et al.* 2002, Marmor & Randlane 2007). Due to specific morphological features — the lack of roots, and acquisition of nutrients and water over the whole body surface (DeWit 1976, Kannukene 1995, Vanderpoorten & Goffinet 2006) — lichens and bryophytes may react more rapidly to airborne pollution than vascular plants. The aim of the current study was to elucidate the vegetation responses to the cement kiln dust pollution gradient on the forest bryophyte *synusiae* level. We also analyzed the reaction of the epiphytic bryophytes not included in our previous paper (Paal *et al.* 2013).

Material and methods

Sample area

We studied the vicinity of the Kunda cement plant located in northern Estonia on the southern coast of the Gulf of Finland (26°32'E, 59°30'N). The area belongs to the mixed-forest subregion of the Atlantic-continental climatic region with an impact of the Baltic Sea (Kaljumäe 1995). The mean annual temperature is +4.9 °C, the lowest mean monthly temperature is in January (–5.7 °C) and the highest in July (+16.2 °C). The annual amount of precipitation is 550–575 mm, the wettest months are July and August. The mean wind speed is 5.2 m s^{–1}, and prevailing winds blow mainly from the south, southwest and west (Annuka 1994), forming the most polluted territory towards the northeast and east from the emission centre (Mandre *et al.* 1992).

The Kunda cement plant was established in 1871 and its surroundings have been affected by cement kiln dust (CKD) emission for almost 140 years. In 1870–1911 annual cement production was 20 000–50 000 tonnes, in 1911–1961 up to 100 000 tonnes. After the reconstruction of the plant in 1962, cement production reached 1.2 million tonnes per year (Noormets & Teedumäe 1995). In the late 1980s and early 1990s, the annual emissions of CKD climbed to about 100 000 tonnes (Estonian Environment 1995). In the winter periods of 1983 and 1985, at a distance of 1.0–1.5 km SW from the plant, the monthly sedimentation intensity on snow was 30–150 g m² (Martin *et al.* 1985). Extrapolating these figures to one year, the amounts of sediments were 0.36–1.8 kg m² (Annuka & Rauk 1992). In the early 1990s, tree trunks, leaves and herbaceous plants at a distance of 2 km from the plant were covered with a crust of CKD, and CKD was still noticeable at a distance of 10 km from the plant (Annuka 1995). After the installation of electrical precipitators in 1996, the dust emission has notably decreased to approximately 160 tonnes in recent years (*see* www.heidelbergcement.com/NR/rdonlyres/19FF5C90-7238-45B1-A722-C353B0518F73/0/Kunda_ENG_0526.pdf).

CKD emitted by the Kunda cement plant contains 40%–50% CaO, 12%–17% SiO₂, 6%–9% K₂O, 4%–8% SO₃, 3%–5% Al₂O₃, 2%–4% MgO, 2.8%–3.2% Fe₂O₃, in small amounts Mn, Zn, Cu and B. pH of CKD in water suspension is 12.3–12.6 (Mandre 2000). In 1994, pH of the melting snow in the vicinity of Kunda exceeded 12, while pH of rainwater at a distance of 0.5 km from the cement plant was 7.1–7.9 and that of snow meltwater 9.1–11.8; in unpolluted areas the corresponding values were 6.1–6.6 and 6.7–7.0 (Mandre *et al.* 2000).

Data collection

The ecosystems being most sensitive and vulnerable to alkaline pollution, besides *Sphagnum*-dominated raised bogs (Karofeld 1996, Paal *et al.* 2010), are boreal pine forests growing on sandy, acidic Humic-Gleyic Podzols (Annuka & Mandre 1995, Reintam *et al.* 2001). In undisturbed conditions, these soils are acidic — pH

of litter and humus horizon is 2.5–4.0; in lower horizons it increases up to 5.0 (Löhmus 2004); they have low buffer capacity and therefore their upper horizons have been shown to react quickly to alkaline pollution (Kokk 1988). Using the Estonian State Forest Management Centre database and forest maps, 20 *Myrtillus* site-type 65–90-year-old pine stands growing on Gleyic Podzols were selected for the study. The stands were located at various distances from the Kunda cement plant, representing the pollution gradients leeward and windward of the plant.

For data collection, a circular sample plot of 0.1 ha was set in the center of a homogeneous stand to minimize edge effects. The tree layer was characterized by the crown closure and the average height (estimated visually), age (estimated by the increment borer), and the tree-trunk basal area (m² ha⁻¹) of every tree species at breast height (1.3 m), which was measured using the Bitte-licht relascope and averaged over 3–5 measurements. The average stem density of shrub species was recorded by counting them in five randomly placed subplots with a radius of 2 m. Young trees of height < 5 m and/or diameter at breast height < 5 cm were also included in the shrub layer. Shrub species outside the randomly situated subplots were recorded with an abundance value 1.

Field and moss layers were described based on the, subsequently averaged, data gathered from 12, 1 × 1 m quadrats in each sample plot; quadrats were located randomly except if they were obviously situated on the border of two microcoenose, in which case they were shifted into the larger microcoenose. In that way we could interpret the data collected in the 1 m² quadrats as representing the synusia (*see* also Gillet 1999). The total cover (%) of the herb and moss layers, as well as the cover (%) of each plant species, were estimated. If some species occurred in a sample plot but were not found in the quadrats, they were assigned 0.01% cover.

Epiphytic bryophytes were sampled on tree bases and trunks to the height of approximately 70 cm, and on decayed logs.

Because airborne alkaline dust sediments have the strongest impact on the forest soil litter horizon (Teras 1984, Kokk 1988, Annuka & Mandre 1995), we gathered a composite sample of soil litter horizon from three randomly selected

pits in each sample plot. In the laboratory, the following soil properties were assessed: (i) pHKCl in 1 N KCl solution, (ii) Mg content from ammonium-acetate solution by Flow Injection Analyser (Page 1982, Ruzicka & Hansen 1981) and Ca content from the same solution by flame photometer (Page 1982), (iii) total nitrogen content according to the Kjeldahl method (Page 1982), (iv) P content by SnCl₂ method and Flow Injection Analyser (Helrich 1990, Ruzicka & Hansen 1981), and K content from the same solution by flame photometer (Helrich 1990), and (v) ash content by ashing sample in 550 °C furnace for 4.5 hours (Ash of Animal Feed (942.05) 1990). All analyses were performed on the fine soil fraction (diameter < 2 mm).

Bryophyte nomenclature follows Ingerpuu and Vellak (1998).

Data processing

Using the earlier pine bark pH estimates (Paal *et al.* 2013) and litter chemistry data (Table 1), the Kunda cement plant surroundings were divided into three pollution zones: I = unpolluted (bark pH ≤ 3.0); II = moderately polluted (bark pH 3.1–4.0); III = heavily polluted (bark pH ≥ 4.1) (Fig. 1). The difference in the mean values

of environmental variables among the pollution zones was tested with one-way ANOVA and Tukey's HSD *post-hoc* test (StatSoft Inc. 2004).

For generalization of relationships among the alkaline pollutants, the litter chemistry data (Ca, K, Mg, Ash, N content and pH) were ordinated by the Principal Component Analysis (PC-ORD ver. 5; McCune & Grace 2002) and the factor scores of the first principal component were interpreted as the compound alkaline pollution factor (CF).

The main gradients of the plant species data were summarized using Detrended Correspondence Analysis (McCune & Grace 2002) with rescaling threshold of 0.1 and 26 segments, axes rescaled from the lowest to the highest score. Percentage of variance in the distance matrix presented by the ordination axes was evaluated by χ^2 -distance. The shared patterns of sample plots, species abundance and environmental variables were portrayed using ordination triplots. The strength of the relationship between environmental variables and ordination axes was evaluated by correlation coefficients.

For cluster analysis, chord distance and flexible β algorithm ($\beta = -0.6$) were applied (McCune & Grace 2002). The chord distance suppresses differences in species total abundance among sample units and compares them accord-

Table 1. Studied environmental variables (mean ± SD) in forests of different pollution zones and significance of differences among zones; pH and concentrations of N, P, K, Ca, Mg and Ash in litter horizon; Closure, Height and Age are respective parameters of the first (highest) tree sublayer. *n* = number of sample plots. Different letters in superscript indicate significant differences as estimated with Tukey's HSD test; *p* = significance level of one-way ANOVA, n.s. = non significant.

Variables	Not polluted (<i>n</i> = 6)	Moderately polluted (<i>n</i> = 4)	Heavily polluted (<i>n</i> = 10)	<i>p</i>
pH	4.0 ^a ± 0.4	6.1 ^b ± 0.5	7.2 ^c ± 0.2	< 0.001
Ca (g kg ⁻¹)	5.5 ^a ± 0.7	17.9 ^a ± 4.0	56.7 ^b ± 29.6	0.001
K (g kg ⁻¹)	0.9 ^a ± 0.1	1.4 ^{ab} ± 0.3	1.85 ^b ± 0.6	0.009
Mg (g kg ⁻¹)	0.5 ^a ± < 0.1	0.6 ^{ab} ± 0.1	1.0 ^b ± 0.4	0.014
N (g kg ⁻¹)	12.9 ^a ± 1.6	11.1 ^{ab} ± 2.8	8.1 ^b ± 4.0	0.031
P (g kg ⁻¹)	1.0 ± 0.1	1.2 ± 0.3	1.2 ± 0.5	n.s.
Ash (g kg ⁻¹)	182.4 ^a ± 158.4	329.2 ^{ab} ± 105.3	375.7 ^b ± 72.1	0.010
Closure	0.5 ^a ± < 0.01	0.7 ^b ± 0.1	0.6 ^{ab} ± 0.1	0.035
Height (m)	23.7 ± 1.0	24.3 ± 1.5	24.6 ± 1.5	n.s.
Age (years)	75 ± 6	78 ± 5	73 ± 9	n.s.
<i>Picea abies</i> basal area (m ² ha ⁻¹)	1.7 ± 0.9	1.0 ± 1.2	1.5 ± 2.7	n.s.
<i>Pinus sylvestris</i> basal area (m ² ha ⁻¹)	22.7 ± 1.8	23.0 ± 2.1	23.4 ± 2.9	n.s.
Basal area of all trees (m ² ha ⁻¹)	24.8 ± 1.5	24.0 ± 2.1	25.1 ± 4.2	n.s.

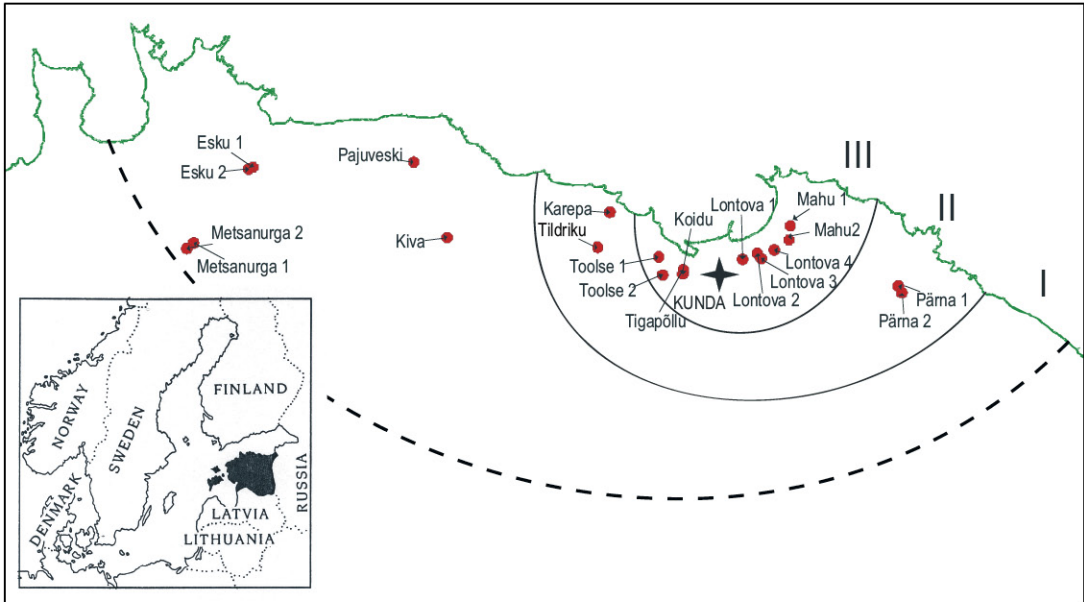


Fig. 1. Location of Kunda cement plant (marked with diamond) and sample plots. I = not polluted zone, II = moderately-polluted zone, III = heavily-polluted zone.

ing to species abundance proportions. Dendrogram “branches” including fewer than five sample quadrats were not considered clusters. To test the differences in species composition among the clusters, we used a non-parametric Multi-Response Permutation Procedure (MRPP; Mielke 1984), with Euclidean distance (McCune & Mefford 1999).

The indicator values of the species in clusters were calculated using the Dufrière and Legendre (1997) method (McCune & Grace 2002). With this method, the relative abundance and relative frequency of each species were calculated for every cluster. Multiplication of these two values, expressed as percentage, yields an indicator value for a particular species in a particular cluster. The statistical significance of the obtained indicator values was evaluated by the Monte Carlo permutation test (4999 runs).

Results

The pH value of the forest litter horizon at a radius of 1.9–2.3 km from the cement plant was 7.1–7.4 and it decreased gradually to 3.6–4.5 at a distance of 30 km. Analogous decreasing gradi-

ent was evident in the content of K, Ca and Mg (Paal *et al.* 2013).

Principal Component Analysis of the litter horizon chemistry data demonstrated a good correlation among several chemical components (Fig. 2). The first principal component extracted 55.5% of the total variance and represents the alkaline pollution gradient. It was most strongly correlated with K and Ca contents (respective correlation coefficients were 0.93 and 0.90), a bit weaker with pH ($r = 0.87$) and Mg content ($r = 0.81$). Scores of sample plots along the second principal component were derived mainly from the litter N content values ($r_{N,II \text{ axis}} = -0.87$); this axis described 22.6% of the total variance. The concentration of P compounds in the litter was mostly reflected by the third principal component, explaining 13.4% of the variance ($r_{P,III \text{ axis}} = 0.94$).

Cluster analysis identified eight bryophyte societies (Table 2), all with significantly different species composition (MRPP, $p < 0.01$) even after Bonferroni correction for multiple comparisons. Though the dominant species of several societies were the same, they had remarkably different abundance proportions in the established societies. Using the dominant and/or indicator species,

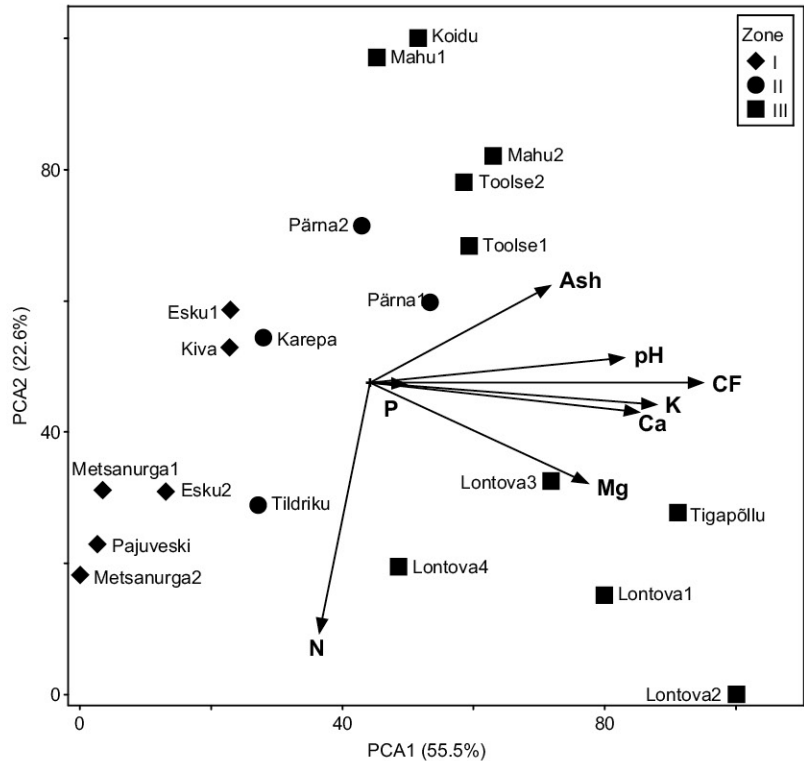


Fig. 2. Ordination of studied forests by Principal Component Analysis using the litter horizon chemistry data. Zone = pollution zone. Symbols: pH = litter horizon pH; Ash, K, Ca, Mg, P and N = values of ash, Ca, K, Mg, P and N content (g kg^{-1}) in litter horizon, respectively; CF = compound alkaline pollution factor.

the societies could be labeled as: *Dicranum polysetum*–*Pleurozium schreberi* soc., *Aulacomnium palustre*–*Hylocomium splendens* soc., *Ptilium crista-castrensis*–*Hylocomium splendens* soc., *Rhytidiadelphus squarrosus*–*Hylocomium splendens* soc., *Climacium dendroides*–*Hylocomium splendens* soc., *Rhodobryum roseum*–*Rhytidiadelphus triquetrus* soc., *Brachythecium rutabulum*–*Rhytidiadelphus triquetrus* soc., *Fissidens dubius*–*Rhytidiadelphus triquetrus* soc.

The ordination triplot (Fig. 3) demonstrates that the sequence of societies in Table 3 follows the pollution gradient described by the calculated compound alkaline pollution factor. Gradient length along the first ordination axis was 3.83 units of species turnover and along the second axis 2.15 units.

In unpolluted forests, the characteristic synusia of the first three societies were dominated by the species common for acidic soils, such as *Pleurozium schreberi*, *Hylocomium splendens*, *Dicranum polysetum* and *Ptilium crista-castrensis* (Table 3). The respective synusia in some microhabitats also included *Sphagnum squarrosum*, *Aulacomnium palustre* and *Poly-*

trichum commune indicating their paludification (Table 2).

In the moderately-polluted zone, the abundances of *Dicranum polysetum* and *Pleurozium schreberi* noticeably decreased and several species (*Aulacomnium androgynum*, *A. palustre*, *Dicranum majus*, *Eurynchium praelongum*, *Pohlia nutans*, *Polytrichum juniperinum*, *P. commune* and *Sphagnum squarrosum*) disappeared altogether. At the same time, in this zone we recorded *Brachythecium rutabulum*, *B. salebrosum*, *Cirriphyllum piliferum*, *Plagiomnium cuspidatum*, *P. rostratum*, *P. undulatum*, *Rhytidiadelphus squarrosus* and *Scleropodium purum* not found in unpolluted forests. The abundance of *Hylocomium splendens* had the highest value in the moderately-polluted zone and in the outermost regions of the heavily-polluted zone — in the *Ptilium crista-castrensis*–*Hylocomium splendens* and *Rhytidiadelphus squarrosus*–*Hylocomium splendens* societies. Two additional societies, *Climacium dendroides*–*Hylocomium splendens* and *Rhodobryum roseum*–*Rhytidiadelphus triquetrus* (Table 3) appear to be related to the alkaline pollution gradient.

Table 2. Centroids of bryophyte societies (mean cover of species) and their indicator species (marked by asterisks; * = $p < 0.05$, ** = $p < 0.01$, *** = $p < 0.001$).

Species	Society							
	<i>Dic pol– Ple sch</i>	<i>Aul pal– Hyl spl</i>	<i>Pti cri– Hyl spl</i>	<i>Rhy squ– Hyl spl</i>	<i>Cli den– Hyl spl</i>	<i>Rho ros– Rhy tri</i>	<i>Bra rut– Rhy tri</i>	<i>Fis dub– Rhy tri</i>
* <i>Aulacomnium palustre</i>	–	0.5**	0.1	–	–	–	–	–
<i>Barbula convoluta</i>	–	–	–	< 0.1	–	–	–	–
<i>Brachythecium erythrorrhizon</i>	–	–	–	–	–	0.4	< 0.1	–
<i>Brachythecium glareosum</i>	–	–	–	–	< 0.1	–	–	–
<i>Brachythecium oedipodium</i>	–	< 0.1	–	–	–	< 0.1	–	–
* <i>Brachythecium rutabulum</i>	–	0.2	0.2	0.2	0.2	0.3	1.2**	0.3
<i>Brachythecium salebrosum</i>	–	0.1	–	0.3	0.3	0.1	–	–
* <i>Bryoerythrophyllum recurvirostre</i>	–	–	–	–	–	–	–	0.2**
<i>Bryum elegans</i>	–	–	< 0.1	–	–	–	< 0.1	0
<i>Campylium calcareum</i>	–	–	–	–	–	0.1	–	–
<i>Campylium stellatum</i> var. <i>protensum</i>	–	–	–	< 0.1	< 0.1	0.1	0.3	0.2
<i>Campylium sommerfeltii</i>	–	–	–	< 0.1	–	–	–	–
<i>Campylium stellatum</i>	–	–	< 0.1	< 0.1	–	–	–	–
<i>Chiloscyphus polyanthos</i>	–	–	–	–	< 0.1	–	–	–
* <i>Cirriphyllum piliferum</i>	–	0.2	0.1	0.4	2.5	2	1.6	4.4***
* <i>Climacium dendroides</i>	–	–	–	–	0.3*	–	–	–
<i>Dicranum majus</i>	0.1	0.1	–	–	–	–	–	–
* <i>Dicranum polysetum</i>	7.7***	2.5	0.7	–	–	< 0.1	–	–
<i>Dicranum scoparium</i>	0.5	0.2	0.1	0.3	–	0.4	–	–
<i>Didymodon fallax</i>	–	–	–	–	–	–	–	0.1
<i>Encalypta streptocarpa</i>	–	–	–	< 0.1	–	–	–	–
* <i>Eurhynchium angustirete</i>	–	–	0.1	0.2	1.0	–	0.9	3.2***
<i>Eurhynchium hians</i>	–	–	–	–	< 0.1	–	< 0.1	0.2
<i>Eurhynchium pulchellum</i>	–	–	–	< 0.1	< 0.1	–	–	–
<i>Fissidens adianthoides</i>	–	–	–	< 0.1	–	–	–	–
* <i>Fissidens dubius</i>	–	–	–	–	–	–	–	0.6***
* <i>Hylacomium splendens</i>	12.5	37.9	63.0	69.4***	44.2	20.9	5.5	0.3
<i>Plagiomnium affine</i>	–	< 0.1	–	–	–	–	–	–
* <i>Plagiomnium cuspidatum</i>	–	–	0.2	0.7	1.4	0.8	0.9	2.1**
* <i>Plagiomnium elatum</i>	–	< 0.1	–	–	–	< 0.1	0.4	1.8***
<i>Plagiomnium ellipticum</i>	–	–	–	0.2	< 0.1	< 0.1	0.2	0.3
<i>Plagiomnium rostratum</i>	–	–	–	–	< 0.1	< 0.1	–	0.1
<i>Plagiomnium undulatum</i>	–	< 0.1	< 0.1	< 0.1	0.2	0.4	0.3	0.6
* <i>Pleurozium schreberi</i>	54.4***	25.5	15.4	1.2	2.0	1.4	–	0.5
<i>Pohlia nutans</i>	–	–	< 0.1	–	–	–	–	–
<i>Polytrichum commune</i>	2.0	1.5	0.1	–	–	–	–	–
<i>Polytrichum juniperinum</i>	–	0.2	0.1	–	–	–	–	–
* <i>Ptilium crista-castrensis</i>	1.0	0.3	1.3*	0.2	–	–	–	–
* <i>Rhodobryum roseum</i>	–	–	–	–	0.1	0.4*	0.2	–
* <i>Rhytiadelphus squarrosus</i>	–	0.2	–	0.6*	–	–	–	–
* <i>Rhytiadelphus triquetrus</i>	–	–	1.0	2.7	15.6	24.8	54.7***	28.5
* <i>Sanionia uncinata</i>	–	< 0.1	–	–	0.1	0.4	0.6*	0.5
* <i>Scleropodium purum</i>	–	0.1	–	< 0.1	–	0.4	< 0.1	3.2***
* <i>Sphagnum squarrosus</i>	2.8***	1.2	0.2	–	–	–	–	–
<i>Thuidium abietinum</i>	–	–	< 0.1	< 0.1	–	–	–	–
* <i>Thuidium philibertii</i>	–	–	< 0.1	0.3	–	–	0.5	1.6***
* <i>Timmia bavarica</i>	–	–	–	–	–	–	0.6	1.6***

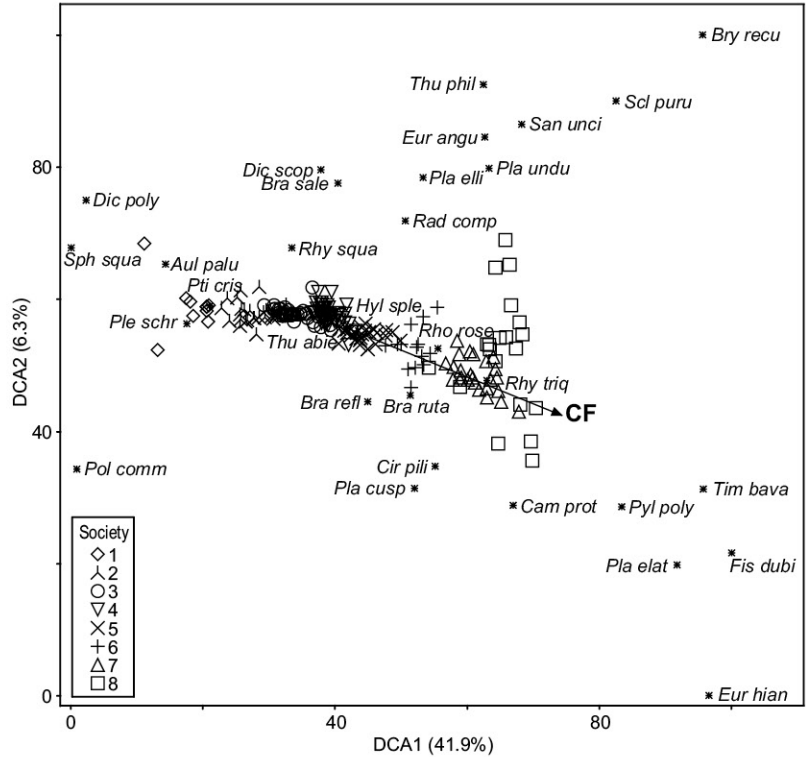


Fig. 3. Detrended Correspondence Analysis ordination triplot of sample quadrats, species recorded at least on three sample quadrats, and compound alkaline pollution factor (CF). For full names of species see Table 2.

Close to the pollution source, the abundances of *Pleurozium schreberi* and *Hylocomium splendens* decreased dramatically, whereas *Brachythecium oedipodium*, *B. velutinum* and *Dicranum polysetum* totally vanished. Instead of species typical of boreal *Myrtillus* site-type forests, calcicolous species or species of boreo-nemoral habitats became common with the increasing pollution load. Significant indicator species for the heavily-polluted zone were *Eurynchium angustirete*, *Plagiommium cuspidatum*, *P. elatum* and *Thuidium philibertii*. In this zone, numerous additional species were documented — *Barbula convoluta*, *Brachythecium erythrorrhizon*, *B. glareosum*, *Bryoerythrophyllum recurvirostre*, *Bryum capillare*, *B. elegans*, *Campylium calcareum*, *C. protensum*, *C. stellatum*, *Didymodon fallax*, *Encalypta streptocarpa*, *Eurhynchium angustirete*, *E. hians*, *E. pulchellum*, *Fissidens adianthoides*, *F. dubius* and *Plagiommium affine*. In the outermost regions of the heavily-polluted zone, we found synusiae of societies presented also in the moderately-polluted zone — *Rhytidiadelphus squarrosus*–*Hylocomium splendens*, *Climacium dendroides*–*Hylocomium*

splendens and *Rhodobryum roseum*–*Rhytidiadelphus triquetrus*; however, *Brachythecium rutabulum*–*Rhytidiadelphus triquetrus* and *Fissidens dubius*–*Rhytidiadelphus triquetrus* societies were most abundant in forests around Lontova, situated in the neighborhood closest to the cement plant (Table 3).

The pollution gradient also affected the species composition of bryophytes growing on tree trunks or on decaying wood (Fig. 4): the correlation coefficient between the compound alkaline pollution factor and the first canonical ordination axis is 0.61. Of the tree layer parameters, the sum of basal area of *Betula* spp. and tree layer closure had the highest correlation ($r = -0.45$ and 0.36 , respectively) with the first ordination axis, and age with the second axis ($r = -0.37$). Gradient length along the first ordination axis is 4.74 units of species turnover and along the second axis 2.76 units.

Species such as *Plagiothecium laetum*, *P. curvifolium*, *Ptilidium pulcherrimum* and *Dicranum montanum* were found within the unpolluted areas (Table 4). With the increasing pollution load, numerous new species colonized these

Table 3. Frequency of bryophyte societies in sample plots in various pollution zones. For full names of species see Table 2.

Zone	Sample plot	Society										Total	
		<i>Dic pol- Ple sch</i>	<i>Aul pal- Hyl spl</i>	<i>Pti cri- Hyl spl</i>	<i>Rhy squ- Hyl spl</i>	<i>Cli den- Hyl spl</i>	<i>Rho ros- Rhy tri</i>	<i>Bra rut- Rhy tri</i>	<i>Fiss dub- Rhy tri</i>				
Not polluted	Metsanurga2	8	3	1									12
Not polluted	Metsanurga1	1	11										12
Not polluted	Esku1	1	2	7	1								11
Not polluted	Esku2		1	9	1								11
Not polluted	Pajuveski			12									12
Not polluted	Kiva		4	7	1								12
Moderate	Pärna1		6	2	2								10
Moderate	Pärna 2				11	1							12
Moderate	Karepa				9	3							12
Moderate	Tiidriku		1	1	2	3			4			1	12
Heavy	Mahu1			4	8								12
Heavy	Mahu2			2	9								12
Heavy	Koidu			3	1	5					1		12
Heavy	Tigapöllu				3	8							12
Heavy	Toolse1			1	4	2							9
Heavy	Toolse2		2		4	1							11
Heavy	Lontova1					1							9
Heavy	Lontova2										7		9
Heavy	Lontova4										9		12
Heavy	Lontova3										3		11
Heavy	Lontova3										1		11
Total		10	30	49	56	24	14	20	25				228

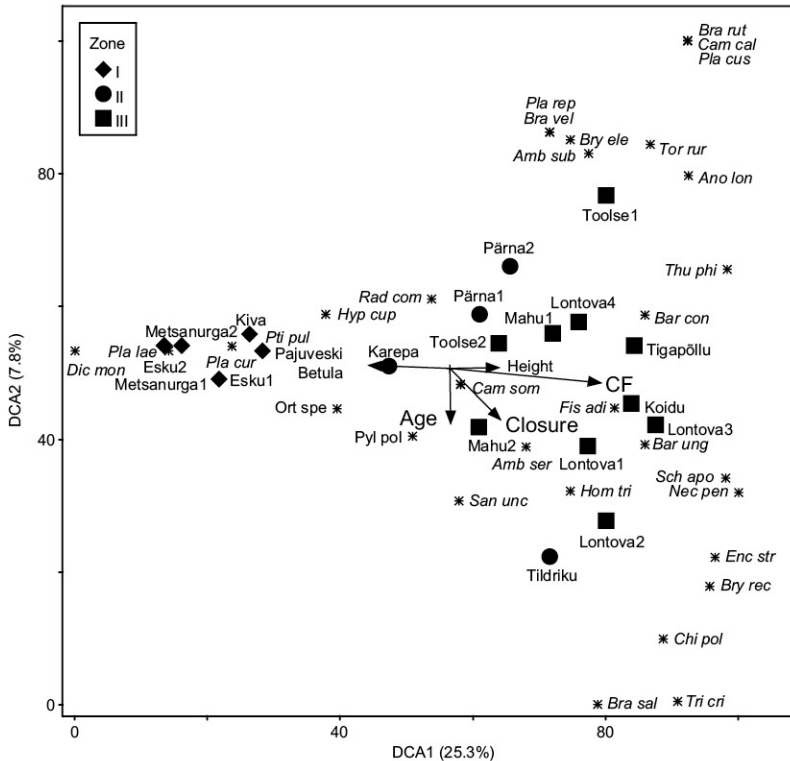


Fig. 4. DCA ordination triplot of sample plots, bryophytes species recorded on trees or on decaying wood, and environmental factors. Notations: *Betula* = basal area of *Betula pendula*, Age = tree layer age, Closure = closure of tree layer crowns, Height = tree layer average height, CF = compound alkaline pollution factor. Abbreviations: *Amb serp* = *Amblystegium serpens*, *Amb subt* = *Amblystegium subtile*, *Ano lon* = *Anomodon longifolius*, *Bar conv* = *Barbula convoluta*, *Bar ung* = *Barbula unguiculata*, *Bra ruta* = *Brachythecium rutabulum*, *Bra sale* = *Brachythecium salebrosum*, *Bra velu* = *Brachythecium velutinum*, *Bry recu* = *Bryoerythrophyllum recurvirostre*, *Bry eleg* = *Bryum elegans*, *Cam calc* = *Campylium calcareum*, *Cam somm* = *Campylium sommerfeltii*, *Chi poly* = *Chiloscyphus polyanthus*, *Dic mon* = *Dicranum montanum*, *Enc stre* = *Encalypta streptocarpa*, *Fis adia* = *Fissidens adianthoides*, *Hom tric* = *Homalia trichomanoides*, *Hyp cupr* = *Hypnum cupressiforme*, *Nec penn* = *Neckera pennata*, *Ort spe* = *Orthotrichum speciosum*, *Pla curv* = *Plagiothecium curvifolium*, *Pla laet* = *Plagiothecium laetum*, *Pla cusp* = *Plagiomnium cuspidatum*, *Pla repe* = *Platygyrium repens*, *Pti pulc* = *Ptilidium pulcherrimum*, *Pyl poly* = *Pylaisia polyantha*, *Rad comp* = *Radula complanata*, *San unci* = *Sanionia uncinata*, *Sch apoc* = *Schistidium apocarpum*, *Thu phil* = *Thuidium philibertii*, *Tor rura* = *Tortula ruralis*, *Tri cris* = *Trichostomum crispulum*.

forests: in moderately-polluted zone we recorded *Radula complanata*, *Pylaisia polyantha*, *Sanionia uncinata*, *Amblystegium serpens*, *Campylium sommerfeltii*, etc. In the heavily-polluted zone, the species composition of epiphytic bryophytes was almost totally changed; it was characterized primarily by *Fissidens adianthoides*, *Tortula ruralis*, *Barbula unguiculata* (Table 4), but there we also recorded *Anomodon longifolius*, *Bryoerythrophyllum recurvirostre*, *Brachythecium glareosum*, *Bryum capillare*, *Campylium calcareum*, *C. stellatum* var. *protensum*, *Didymodon fallax*, *Encalypta streptocarpa*, *Fissidens dubius*,

Neckera pennata, *Plagiomnium cuspidatum*, *Schistidium apocarpum*, *Thuidium philibertii*, *Timmia bavarica* and *Trichostomum crispulum* (Fig. 4).

Discussion

When deposited, the airborne CKD pollution will induce alkalization of the litter horizon, expressed in remarkable increase of its pH, and decrease of exchangeable and hydrolytical acidity (Teras 1984, Kokk 1988, 1992). Moreover,

the alkaline dusty deposition leads to the damage of mycorrhizae in soil (Haapala *et al.* 1996a), to increased microbial degradation of humus and higher content of soluble organic compounds which can form soluble complexes of heavy metals (Haapala *et al.* 2001), and to decreased buffer capacity of soils on calcareous rocks (Reintam *et al.* 2001). As we noted in our previous paper (Paal *et al.* 2013), despite marked reduction of alkaline emissions during the last 15 years, the Ca content of the litter horizon in the heavily-polluted zone still exceeded that of the unpolluted areas ten-fold; the Mg and K contents were doubled. The increasing trend in pH of pine bark was also clearly parallel to the CKD gradient. Pine bark pH had a value of 2.4–2.7 in the unpolluted, distant stands; at the distances of 7 km upwind and 10 km downwind from the Kunda cement plant it varied between 3.2 and 3.7, whereas in stands situated less than 2 km upwind and 2–5 km downwind the respective values were 6.1–6.8. In the town of Kunda, in the neighborhood closest to the plant, pine bark pH reaches value 8.1.

Kannukene (1995) reported that, in the 1990s, the epiphytic bryoflora in the heavily-polluted vicinity of the Kunda cement plant was remarkably species rich. It included calcicolous *Amblystegium serpens*, *Brachythecium glareosum*, *Bryoerythrophyllum recurvirostre*, *Didymodon fallax*, *Gymnostomum calcareum*, *Neckera complanata*, *Tortula ruralis* var. *calcicola*, etc., and pioneer species with a wide ecological amplitude as regards substrate pH and nutrient

content, e.g. *Barbula convoluta*, *B. unguiculata* and *Bryum argenteum*. Martin *et al.* (1990) and Liblik *et al.* (2003) highlighted *Ceratodon purpureus*, *Ortotrichum obtusifolium* and *Tortula ruralis* as occurring with high frequency in the polluted area. Moreover, species indifferent to alkaline pollution, such as *Hypnum cupressiforme*, *Ortotrichum speciosum* and *Pylaisia polyantha*, were common. *Drepanocladus uncinatus*, *Radula complanata* and *Homalia trichomanoides* occurred on tree bark in the moderately-polluted zone. Our study confirmed the consistently high species richness of both epiphytic bryophytes and bryophytes on decaying wood in the heavily-polluted zone. Besides the epiphytic species noted above, *Amblystegium subtile*, *Anomodon longifolius*, *Bryum flaccidum*, *Neckera pennata*, *Schistidium apocarpum* and *Trichostomum crispulum* appeared to be confined to the most polluted area. Several species among them (e.g. *Amblystegium serpens*, *Ceratodon purpureus*, *Bryoerythrophyllum recurvirostre*, *Neckera pennata*, *Schistidium apocarpum*, *Barbula convoluta* and *B. unguiculata*) showed high tolerance to alkalinity elsewhere, often occurring on limestone outcrops or on concrete substrates (Giesy 1957, Ingerpuu & Vellak 1998). In contrast, species usually growing on stem bases (e.g. *Plagiothecium laetum*, *P. curvifolium* and *Dicranum montanum*) were absent from the heavily-polluted zone. Absence of those species from areas treated with lime was also reported by Dulière *et al.* (1999). Those species presumably have a much narrower range of pH tolerance.

Table 4. Indicator species of epiphytic bryophytes for different pollution zones, their indicator values and relative frequencies. Symbols: *p* = significance level by the Monte Carlo permutation test, n.p. = not polluted.

Species	<i>p</i>	Zone	Indicator value			Relative frequency		
			not polluted	moderate	heavy	not polluted	moderate	heavy
<i>Plagiothecium laetum</i>	0.001	not polluted	83	0	0	100	0	0
<i>Dicranum montanum</i>	0.003	not polluted	67	0	0	100	0	0
<i>Ptilidium pulcherrimum</i>	0.007	not polluted	64	0	0	83	25	0
<i>Amblystegium serpens</i>	0.018	moderate	0	50	50	0	100	100
<i>Pylaisia polyantha</i>	0.022	moderate	1	48	39	17	100	90
<i>Radula complanata</i>	0.030	moderate	0	49	14	0	75	40
<i>Fissidens adianthoides</i>	0.002	heavy	0	0	80	0	0	100
<i>Tortula ruralis</i>	0.006	heavy	0	0	70	0	0	100
<i>Barbula unguiculata</i>	0.013	heavy	0	0	60	0	0	100

Though bryophytes are more sensitive to changes in the environment than vascular species (Barkman 1968, Carlberg *et al.* 1983, Dierschke 1994), only a few ground-dwelling bryophyte species are considered to be indicative of alkaline dust pollution. According to Stravinskienė *et al.* (2004) and Stakėnas and Sujetovinė (2005) *Campylium stellatum* and *C. sommerfeltii* belong among them. We recorded these species and *C. calcareum* only in forests within the heavily-polluted zone; however, they were not identified as indicator species of alkaline pollution because their abundance was very low. According to Sujetovinė (2008), *Calliergonella cuspidata*, *Plagiogonium affine*, *P. cuspidatum* and *Eurynchium angustirete* also occur in polluted areas. Except *Calliergonella cuspidata*, the other species were typical of the heavily-polluted zone in forests around the Kunda cement plant. Moreover, Kannukene (1995) pointed out dominance of calcicolous *Timmia bavarica* on all substrates in the zone of very high cement dust load. Our results support the suggestion that this species is a significant indicator of substrate alkalization.

As previously noted (Paal *et al.* 2013), many bryophyte species recorded in the heavily-polluted zone had insufficient abundance to serve as indicator species; therefore, if we perform analyses at the species level, these species should form an indicator-species complex. Analysis at the synusia level gives a more stable result. The relatively consistent recurrence of synusiae of certain societies in the same zone of the pollution gradient provides a more reliable interpretation of the process of moss layer succession.

In the moss layer of the Estonian *Vaccinium* site type, discontinuity rather than continuity was prevalent (Paal 1994). This was presumably a result of spatially discrete microhabitat conditions (Barkman 1968, 1973, Norin 1979, Dierschke 1994), different ways/strategies of species' distribution (Wilmanns 1989) and uneven probability of species' co-occurrence, i.e. assembly rules (*see* Götzenberger *et al.* 2012). Therefore, changes in plant cover will not occur as a gradient but as a mosaic of discrete assemblages (Razumovskii 1981, Yastrebov 1991, Paal 1994). Patterns occur at the scale of synusiae (Korchagin 1976, Minarski & Daniēls 2006) or (for field and moss layers together) at the scale of

microcoenoses (Barkman 1968, Mirkin 1986). In boreal forests that corresponds in metric scale to 0.1–10 m², e.g. in mixed *Vaccinium* site type *Picea abies*–*Pinus sylvestris* stands in southern Estonia the diameter of moss synusiae varies from 0.3 to 1.4 m (0.1–2.1 m²) (Paal 1994).

Komarova (1993) stated that, after fires, three main processes occur in the structural changes of field layer synusiae of *Pinus sibirica* forests in the Sikhote-Alin mountains: (i) some types of synusiae vanish, (ii) synusiae of some other types are partly replaced by the synusiae already existing in the community, and (iii) synusiae of new types previously not present in the community emerge due to the disappearance of previous species and introduction of new ones. Our results found evidence of all these changes in species composition in the bryophyte synusiae, and their typological structure changed significantly along the alkaline pollution gradient. From this, we conclude that *Rhytidiadelphus squarrosus*–*Hylocomium splendens*, *Climacium dendroides*–*Hylocomium splendens* and *Rhodobryum roseum*–*Rhytidiadelphus triquetrus* societies characterize the areas of moderate pollution with pine bark pH 3.1–4.0 and litter horizon pH 5.7–6.8, whereas *Brachythecium rutabulum*–*Rhytidiadelphus triquetrus* and *Fissidens dubius*–*Rhytidiadelphus triquetrus* societies indicate forests affected by heavy alkaline pollution where pH of pine bark is ≥ 4.1 and that of litter horizon ≥ 6.8 .

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