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## IMPACT OF OIL SHALE FLY ASH EMITTED FROM A POWER PLANT ON RADIAL GROWTH OF SCOTS PINE IN NORTH-EAST ESTONIA

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*Response of radial growth of Pinus sylvestris to pollution caused by oil shale fly ash deposition on forest stands in the vicinity of the Baltic Power Plant, North-East Estonia, was studied. Multivariate analysis indicated that oil shale fly ash may promote annual radial growth of Scots pine. The coefficients for emission predictors were positive in all computed regression equations. The favourable impact of oil shale fly ash on the radial growth of younger trees started to decrease after 3-4 years. In the case of older trees the impact of oil shale fly ash emissions lasted 5-6 years.*

### Introduction

Several methods can be used for studying changes in the environment of tree growth. The method of dendrochronology has become widespread as a tool in environmental and ecological research. Forest stand dynamics and ecological history, particularly forest damages caused by various pollutants have been issues of particular interest in recent decades [1-5]. Coupling tree-ring chronologies, pollution history and meteorological data provides a valuable method for understanding forest responses to spatial and temporal changes in the environment of tree growth.

In the industrial region in North-East Estonia alkaline dust pollution from oil shale processing, industry of building materials and power engineering requires special attention as here dry deposition constitutes on the average as much as 45-50 % of total air pollution [6]. The effect of dust emitted by the industry of building materials on plants may be neutral, stimulating or toxic

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depending on the type, concentration of components, level of deposition and meteorological conditions [7, 8]. Some authors have recommended cement-kiln dust as a fertilizer or neutralizer of acid soils [9, 10]. Others have shown its deleterious effect on vegetation [11, 12]. The poor understanding of the effects of various pollutants, especially for relatively low doses, makes the identification of the pollution signal in tree rings difficult and the results controversial [13].

A variety of tree species have been studied in their response to dust. A review of literature reporting such research was presented by Farmer [14]. However, only a few studies directly concerned with coniferous trees are listed. The goal of the present study was to provide information about the response of radial growth of Scots pine to pollution caused by oil shale fly ash deposition on forest stands.

### Study Area and Study Sites

This study was carried out in the northeastern industrial region of Estonia. The topography of the study area is characterized by old postglacial sea dunes. The mean annual air temperature and total annual precipitation are +4.4 °C and 640 mm, respectively. The prevalent winds blow from the southwestern at an annual mean speed of 4.4 m sec<sup>-1</sup> [15]. The dominant forest formations are dry boreal Scots pine (*Pinus sylvestris* L.) stands of varying age.

The main sources of atmospheric pollution in this area are the oil-shale-fired Baltic and Estonian Power Plants, which started operation in 1959 and 1969, respectively. Emissions reached their maximum in 1990, showing a decreasing trend afterwards [15]. Pollutants emitted from the plants consist of hazardous gaseous components (SO<sub>2</sub>, NO<sub>x</sub>) and oil shale fly ash, which contains various phytotoxic elements [16].

In the study area two single-species Scots pine forest stands were chosen as study sites. The stands on these study sites are defined as *Vaccinium*-site type [17]. Study sites Narva-1 and Narva-2 are located in Narva-Jõesuu, approximately 11 km north from the Baltic Power Plant and 22 km northeast from the Estonian Power Plant. Thus the stands at the study sites are directly influenced by the emissions of the Baltic Power Plant.

The stand at the study site Narva-1 is well stocked and 75 years of age. The competition for light and root competition among trees are strong. The study site Narva-2 is represented by a stand where the age of canopy dominants is approximately 150 years, and it has pine undergrowth of varying age. The competition for light between dominant trees is not strong, but the root competition due to undergrowth may be a significant factor affecting the tree growth. No disturbances and management history are known for any sampled stands.

## Methods

### Sampling and Measurements

At each of the two study sites 20 dominant or co-dominant trees were sampled for analysis of radial growth. Lack of damage or defect was considered in sample tree selection. Each sample tree was cored at breast height on the southern and northern sides of the bole using a 4.3-mm increment corer. In laboratory each core, when dry, was mounted onto a grooved holder and the surface of the cores was cleaned and finished up using sandpaper. The cores were crossdated with each other by regional pointer years to identify missing or false rings. The widths of tree rings on cores were measured to the nearest 0.01 mm with the Metronics tree-ring measuring system. Crossdating quality was assessed using the COFECHA program available in the Dendrochronology Program Library (DPL) version 2.1 [18]. The trees with cores which were impossible to crossdate or poorly correlated with others were eliminated from further analysis.

### Statistical Analysis

For estimating the relationships between the radial growth of pine stands and the oil shale fly ash emissions from the power plants two steps of the statistical analysis were applied. At the first step the response of the radial growth to the impact of different climatic factors was studied. For the response analysis the program RESPO, included in the DPL, was used. Relationships between the tree growth and climate were estimated comparing the site chronologies with mean monthly temperatures and total precipitation from the previous June to the current July throughout the period 1960–1991. The climatic data of the previous year were included in the analysis because the climate of the preceding growth session influences the tree growth in the current year [19]. Because the growth rate in the current year may be dependent on the prior growth, the ring-width indices of the three preceding years were included in the pool of predictors as well. As a result of the analysis a multiple regression was computed. The regression equation describes the dependence of the radial growth on the most significant climatic predictors.

At the second step multivariate analysis of the relationships between oil shale fly ash emissions and radial growth of trees was used. The data of oil shale fly ash emissions were included in the regression equation along with the selected climatic factors and prior growth data using the Stepwise Variable Selection procedure in the statistical software program STATGRAPHICS. If the value of the coefficient of determination ( $R^2$ ) increased after the inclusion of the emission data, and the value of the regression coefficient of the emission predictor was above zero, the impact of oil shale fly ash on the growth was considered positive. The decreasing value of  $R^2$  indicates the depreciation of the relative importance of oil shale fly ash on the growth.

The series of tree-ring widths contain normally a considerable amount of non-climatic signals that may include either a biological growth trend, tree disturbance signals, or both [19, 20]. In tree-ring analysis, the procedure of removing non-climatic variance from tree-ring series is known as standardization (or detrending). Standardization transforms ring widths into new series (chronologies) of relative tree-ring indices. This is achieved by fitting an appropriate curve to a ring-width series and dividing each measured ring width by its expected value. In an open canopy stand with minimal competition among trees the growth trend of trees can be adequately modelled by the negative exponential function applied in some cases in combination with linear regression lines.

In dense forests the competition between trees is great and individual trees face different unpredictable disturbances throughout the life. Growth curves of trees are complex and vary between trees. In such cases the application of stochastic methods of standardization (cubic-smoothing splines, double detrending) is appropriate [21].

In the present work standardization was accomplished with the ARSTAN program version 6.04P in DPL. The standard (ST) and ARSTAN versions of final tree-ring chronologies were produced. The standard version of chronologies includes a large portion of the impact of the prior growth (persistence) on the ring width of the growth year. The ARSTAN chronologies contain the persistence that is common and synchronous among a large portion of the tree-ring series from the site, without including that found in only one or very few series [22]. The ARSTAN chronologies contain the strongest climatic signal possible.

## Climate and Emission Data

The monthly average temperatures and monthly total precipitation data were obtained from the Narva meteorological station of the Estonian Institute of Meteorology and Hydrology. The emission data for the Baltic Power Plant were provided by the AS *Eesti Energia* (*Estonian Energy Ltd*).

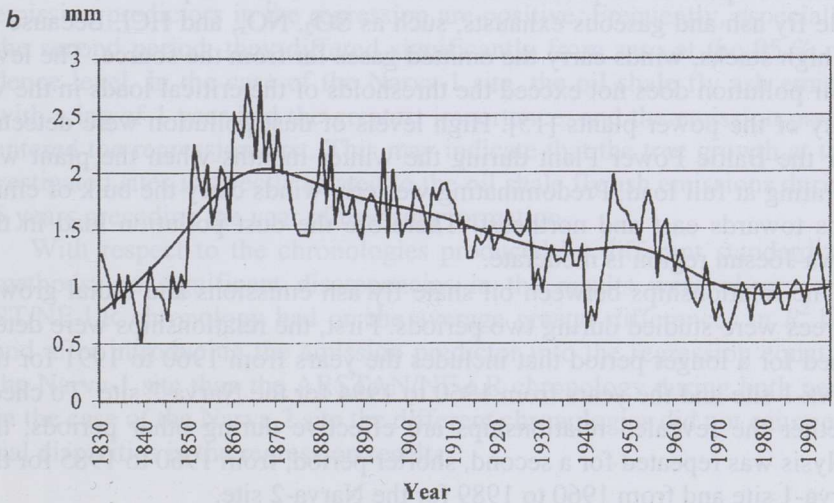
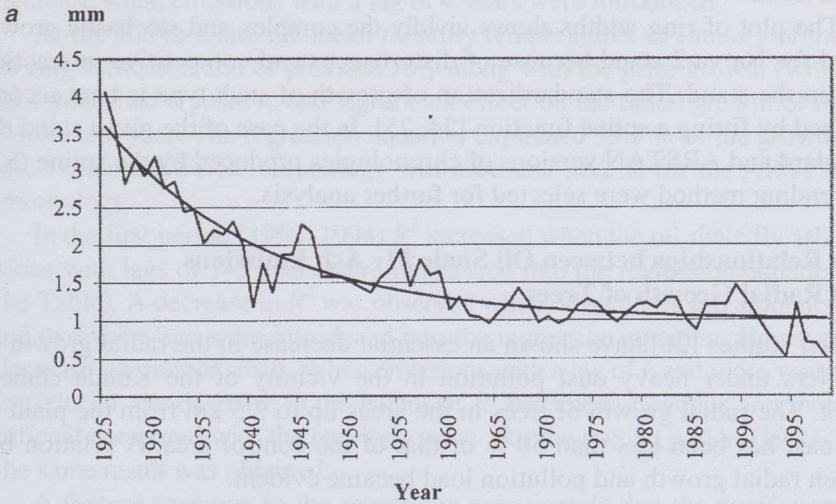
## Results and Discussion

### Radial Growth of the Stands and Selection of the Standardization Method

**Narva-1.** The lack of abrupt long-term changes in tree-ring widths (Figure) suggests that the growth of trees is limited for the most part by natural environmental factors. The wide innermost tree-rings indicate that initially the trees grew without competition. The further growth curve is best described by fitting a negative exponential equation. As mentioned above, such growth-curve type is specific for stands with minimum competition among trees and without strong external influences. For this reason the standard and ARSTAN versions of chronologies produced by the negative exponential (if it fails, a linear regression line) detrending (NE-LR) were

it fails, a linear regression line) detrending (NE-LR) were selected for further analysis.

Against the background of the general decrease in radial growth, the wide tree-rings in years 1945, 1955, 1967, and 1989 may be distinguished. The growth was suppressed in years 1940, 1964, 1985, and 1994. The extremely narrow tree-ring in 1940, which is visible in many pine tree-ring chronologies in Estonia and in neighbouring areas [23], can be attributed to the synergetic effect of the very dry summer in 1939 and the frosty winter of 1939–1940. On the diagram an upward trend of the radial growth from 1970 until 1989 followed by a sharp decline during the next years can be observed.



Radial growth and smoothing curves (negative exponential for the Narva-1 site (a) and spline for the Narva-2 (b) site) of Scots pine stands at the sites Narva-1 and Narva-2

**Narva-2.** A series of narrow tree-rings up to 1840 suggests that young trees were subjected to competition stress caused by the overwood. The increase in radial growth starting from 1850 is obviously a result of a large-scale thinning of the upper storey. The maximum value of the radial growth was achieved in 1863–1865. As a result of the increasing competition among trees and the increasing age the radial growth decreased gradually up to a second minimum in 1940–1942. After this minimum a sharp increase in the radial increment in 1943–1946 due to favourable meteorological conditions of these years can be observed in the plot of ring widths. Starting from 1946 until 1971 radial growth showed a tendency of declining followed by an upward trend.

The plot of ring widths shows vividly the complex and stochastic growth trend for Narva-2 stand because of disturbances and competitive interaction within the stand. The standardization of growth of such type is best accomplished by fitting a spline function [24, 25]. In the case of the given stand the standard and ARSTAN versions of chronologies produced by the spline (SP) detrending method were selected for further analysis.

### **The Relationships between Oil Shale Fly Ash Emissions and Radial Growth of Trees**

Earlier studies [26] have shown an essential decrease in the radial growth of conifers under heavy dust pollution in the vicinity of the Kunda cement plant. The radial growth of trees in the areas up to 2.5 km from the plant to the east has been less than 50 % of that of the control area. A relation between radial growth and pollution load became evident.

The complex of air pollutants emitted from the power plants contains oil shale fly ash and gaseous exhausts, such as  $\text{SO}_2$ ,  $\text{NO}_x$ , and HCl. Because of the high stacks, winds carry the emitted gases far from the source. The level of air pollution does not exceed the thresholds of the critical loads in the vicinity of the power plants [15]. High levels of dust pollution were detected near the Baltic Power Plant during the winter months when the plant was operating at full load. Predominating westerly winds carry the bulk of emissions towards east and northeast. Therefore the dust pollution load in the Narva-Jõesuu region is moderate.

The relationships between oil shale fly ash emissions and radial growth of trees were studied during two periods. First, the relationships were determined for a longer period that includes the years from 1960 to 1991 for the Narva-1 site and the years from 1960 to 1994 for the Narva-2 site. To check whether the revealed relationships are effective during other periods, the analysis was repeated for a second, shorter period, from 1960 to 1985 for the Narva-1 site and from 1960 to 1989 for the Narva-2 site.

The results of the response function analysis revealed significant positive relationships between the radial growth of trees at the Narva-1 site and the mean monthly temperatures of February of the year of ring formation. Also total precipitation in previous August and the prior growth (with a lag of

1 year) had a positive and significant effect on the radial growth of trees in the current year at this site. These factors were introduced as predictors into the regression equation. The percentage of the growth variance explained by the equation was 30.9 % when the ST/NE-LR chronology was used as a growth variable and 26.0 % in the case of the ARSTAN/NE-LR chronology.

In the first period (1960–1991) the coefficient of determination ( $R^2$ ) increased when the oil shale fly ash emissions with lags of 1–4 years were introduced into the regression equation (Table). When emissions with a lag of 5 years were introduced, the values of  $R^2$  started to decrease compared with the previous value. In the second period (1960–1985) the values of  $R^2$  started to decrease when emissions with a lag of 4 years were introduced.

At the Narva-2 site the mean monthly temperatures of January of the year of ring formation and of previous July along with the prior growth (with a lag of 1 year) had a positive and significant effect on the radial growth of trees in the current year. The regression equation explained 35.4 % of the growth variance when the ST/SP chronology was used and 36.2 % for the ARSTAN/SP chronology.

In the first period (1960–1994)  $R^2$  increased when the oil shale fly ash emissions with lags of 1–5 years were introduced into the regression equation (see the Table). A decrease in  $R^2$  was observed when oil shale fly ash amounts emitted two years ago were introduced into the regression equation. However, this decrease was rather small. When emissions with a lag of 6 and more years were introduced, the decrease in the values of  $R^2$  became continuous and more significant compared with the previous value. For the second period (1960–1989) the same result was obtained.

A feature common to the regression equations is that the coefficients for emission predictors in the regression are positive. Frequently, especially for the second period, they differed significantly from zero at the 95 % confidence level. In the case of the Narva-1 site, the oil shale fly ash emissions with a lag of 1 year had the greatest importance, and the emission predictor entered the regression first. This may indicate that the tree growth at the investigated sites is directly related to the oil shale fly ash emissions during 3–5 years preceding the year of tree-ring formation.

With respect to the chronologies produced by different standardization methods, no significant discrepancies in the results were observed. The ST/NE-LR chronology had on the average greater differences in  $R^2$  before and after introducing the emission predictor into the regression equation at the Narva-1 site than the ARSTAN/NE-LR chronology during both periods. In the case of the Narva-2 site the different chronologies did not cause essential disparities in the regression results.

Results of Regression Analysis for Radial Growth versus Oil Shale Fly Ash Emissions during Two Periods.  
 First Period: 1960–1991 for the Site Narva-1 and 1960–1994 for the Site Narva-2;  
 Second Period: 1960–1985 for the Site Narva-1 and 1960–1989 for the Site Narva-2

Standardization method	First period			Second period				
	Emissions in regression (lag in years)	Coefficient for the emission predictor in regression	$R^2$ of regression		Emissions in regression (lag in years)	Coefficient for the emission predictor in regression	$R^2$ of regression	
			Before introducing the emission predictor	After introducing the emission predictor			Before introducing the emission predictor	After introducing the emission predictor
ST/NE-LR	1	0.00177*	0.2326	0.3279	1	0.002062*	0	0.1115
	2	0.001262	0.3075	0.3668	2	0.001486	0.2377	0.3001
	3	0.001603	0.2408	0.3476	3	0.001681*	0.2537	0.3451
	4	0.001064	0.3549	0.3864	4	0.000792	0.3419	0.3379
	5	0.000442	0.3712	0.3501	5	0.000043	0.3819	0.3477
	6	0.000262	0.3666	0.3402				
ARSTAN/NE-LR	1	0.001618*	0.2636	0.3475	1	0.001935*	0	0.0959
	2	0.001262	0.2597	0.3014	2	0.001491*	0.1687	0.2397
	3	0.001487	0.2638	0.3227	3	0.001546*	0.1739	0.2511
	4	0.000901	0.3062	0.3132	4	0.000697	0.2621	0.2506
	5	0.000336	0.3124	0.2892	5	0.000019	0.2994	0.2605
	6	0.000402	0.3133	0.2888				

\*  $p < 0.05$



Results of Regression Analysis for Radial Growth versus Oil Shale Fly Ash Emissions during Two Periods.  
 First Period: 1960–1991 for the Site Narva-1 and 1960–1994 for the Site Narva-2;  
 Second Period: 1960–1985 for the Site Narva-1 and 1960–1989 for the Site Narva-2 (end)

Standardization method	First period				Second period			
	Emissions in regression (lag in years)	Coefficient for the emission predictor in regression	$R^2$ of regression		Emissions in regression (lag in years)	Coefficient for the emission predictor in regression	$R^2$ of regression	
			Before introducing the emission predictor	After introducing the emission predictor			Before introducing the emission predictor	After introducing the emission predictor
ST/SP	1	0.001442	0.3536	0.3773	1	0.001574*	0.3241	0.3553
	2	0.000892	0.3756	0.3713	2	0.001315	0.3538	0.3658
	3	0.001051	0.3714	0.3732	3	0.001293	0.3471	0.3556
	4	0.001153	0.3687	0.3773	4	0.001578	0.3205	0.3484
	5	0.001417	0.3572	0.3724	5	0.001418	0.3505	0.3589
	6	0.00115	0.3731	0.3714	6	0.001238	0.3541	0.3519
	7	0.001103	0.3542	0.3488	7	0.001061	0.3348	0.3238
ARSTAN/SP	1	0.001561	0.3541	0.3689	1	0.001632	0.3211	0.3379
	2	0.000995	0.3851	0.3844	2	0.001367	0.3587	0.3734
	3	0.001212	0.3832	0.3921	3	0.001397	0.3411	0.3571
	4	0.001173	0.3829	0.3879	4	0.001556	0.3419	0.3665
	5	0.001473	0.3751	0.3925	5	0.001449	0.3514	0.3626
	6	0.001123	0.3819	0.3794	6	0.001188	0.3595	0.3553
	7	0.001051	0.3578	0.3508	7	0.000998	0.3342	0.3212

\*  $p < 0.05$

Dust can affect plants in different ways. Dust and aerosols coming from the atmosphere to the forest ecosystem are deposited mostly on leaves. If an arid period during the growth season lasts for a long time, a crust forms on needles under a high dust pollution load. This dust can block stomata, causing thus disorders in physiological processes connected with assimilation in leaves. The results of an experiment with Scots pine seedlings demonstrated a 25-% lower starch content in trees with incrustated needles and the inhibition of the translocation of carbohydrates into roots and the stem [6]. Therefore, dust deposition leads generally to growth reduction.

Another way dust affects plants goes *via* changes in soil chemistry. Accumulation of additional amounts of Ca, Mg, K and Na causes alkalization and an increase in the pH values of soils affected by dust [27–29]. This complicates mineral nutrition and disbalances the content of micro- and macro-elements in the organism, leading finally to a decrease in bioproduction [26, 30]. On the other hand, the nutrient pool of *Vaccinium*-site type sandy soils is generally poor and depends much on the depth of the more fertile moraine layer [17]. In dune areas the moraine layer is relatively deep-seated and therefore the supply of trees with nutrients is problematic. Studies have shown that in areas where the dustiness of the air is high, the deposited inorganic particles and aerosols may be an additional source of nutrients for ecosystems [31].

Most studies on dust effects have revealed a deleterious effect of dust on various tree species. Usually the physical injuries to leaves, chlorotic needles, cell destruction and reduced growth were observed [32–34]. In Estonia a strong negative effect of cement dust pollution on the radial growth of conifers (less than 50 % of the control) was observed near a cement plant after the 1950s, when the production increased sharply [26].

Dust deposition has also been found to have a positive effect on the growth of conifers. Havas and Huttunen [1] reported a clear increase in radial growth of Scots pine of different age in forests surrounding the fertilizer plant of *Typpi Oy* in Oulu. The favourable impact of dust on the volume increment of conifers in the vicinity of a cement plant in Latvia was shown by Skudra [35]. The dependence of the pollution–growth relationships on prevailing winds, distance from sources and chemical composition of pollutants, however, was admitted by authors.

The results of the statistical analyses carried out in the given study revealed a positive impact of oil shale fly ash emissions on radial growth of pines at the both investigated stands. Preliminary analysis did not show any essential improvement of climatic conditions during the period investigated. The average annual mean temperature for the period 1960–1994 turned out to be even slightly lower than the long-term average. An increase in annual total precipitation during this period was observed, however. Commonly soil moisture is not a factor limiting the growth of pines at this site type [36]. The increase in the radial growth starting from 1970, noticeable on the diagrams, may have been caused by improved nutrition conditions. In the study area

the nutrient deficiency may be partially covered by the oil shale fly ash deposition, which is quite rich in different nutrient elements (e.g. K, Ca, Mn, Mg, microelements) [16].

Though the obtained results coincide with the arguments about a positive impact of low dust concentrations upon the tree growth, the author believes that a replication of studies on different sites and at different dust loads might yield more reliable and precise results.

## Conclusions

Multivariate analysis of the relationships between oil shale fly ash emissions and radial growth of trees indicated that annual radial growth of Scots pines might be influenced positively by oil shale fly ash. The coefficients for emission predictors were positive in all computed regression equations. The positive impact of oil shale fly ash on the radial growth of younger trees started to decrease after 3-4 years. In the case of the older trees the impact of oil shale fly ash emissions lasts 5-6 years.

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