THE CONTENT OF HEAVY METALS
IN THE WOOD OF HEALTHY AND DYING OAK TREES
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Abstract. The total content of mineral substances (ash) and heavy metals (Al, Cd, Cr, Cu, Fe, Mn, Pb) in the wood of healthy and dying oak trees from different regions of Poland was subject to analysis. The material for analysis came from the 12 Forest Districts in which the main source of oak wood in our country is located. Two trees healthy and dying of the same age were cut in each stand showing symptoms of a decline trees in the past and at present. Wood samples cut out of the outer heartwood at the butt-end of the trunk of oak trees aged 60-155 were used for analysis. The content of mineral substances in the wood of healthy and dying trees was at a similar level. No statistically significant difference in the content of each of the analysed elements was found in the wood of healthy and dying trees.

Key words: heavy metals, health condition, pedunculate oak, sessile oak, oak wood, Poland, tree vitality, forest decline

INTRODUCTION

In our geographic latitude, oaks are the main forest tree species. Oak stands play an important ecological and productive role supplying valuable wood assortments.

The process of increased decline of broadleaved forests, especially oak forests, and excessive natural thinning of trees has been observed in Poland and Europe since the beginning of the 1980s. Oak decline is a complex disease in terms of both, aetiology and symptomatology. For many years, attempts have been made to find the causes of oak decline and explain the development of this phenomenon from different angles. The prevailing opinions are that the nature of the disease is complex and caused by many factors, and that these predisposing, initiating and contributing. Factors operate in oak

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ecosystems concurrently or in a specific sequence. The stress factors of abiotic, and biotic origin give rise to unfavourable physiological and physicochemical changes causing that the natural resistance of a tree is drastically reduced and the tree, being unable to compete with other individuals, dies [Przybył 1995, Siwecki and Ufnalski 1998, Recent advances... 2000, Sierota 2001, Thomas et al. 2002, Protection... 2004, Oszako 2007].

Among many factors responsible for the weakening and decline of many tree species, anthropogenic contamination of the environment is considered to be most important. Numerous studies were conducted on the impact of environmental pollution on the macro- and microelement concentrations in different tree parts. The mean content of mineral substances, calculated per dry mass of wood, under Poland's climatic conditions oscillates between 0.3 and 1.0%. Although the quantity of mineral substances in wood is relatively small, many of them are of great importance for tree growth. The content of mineral substances in individual parts of a tree increases in the following sequence: trunk, branches, roots, shoots, fine roots, bark, foliage. The content of mineral compounds in wood also depends on the species, growth conditions (soil, climate, environment) or felling time [Prosiński 1984, Fengel and Wegener 1989].

There is a scarce number of studies dealing with a comparison of heavy metal concentrations in the stem wood of oak trees growing under the same environmental conditions but varying in healthiness level.

The performed studies are aimed at determining the content of heavy metals in oak wood in relation to the health condition of trees. The oak wood samples came from stands in those Forest Districts where oak trees showed symptoms of decline and excessive self-thinning over the past two decades. A pair of trees ("healthy" – "dying") of the same age was selected from the same stand to reduce the impact of the environment on the examined parameter. When such a tree pair could not be found within one stand, it was selected from different stands with comparable natural/forest characteristics. The material for analysis was collected from stands from different regions of Poland, which may point to the possibility of spatial (geographic) variability of a given parameter of oak wood. The greatest number of wood samples taken for analysis are represented by the pedunculate oak. Only one site (Świerczyna Forest District) is represented by the sessile oak.

MATERIAL AND METHODS

The material for the studies was collected from 13 stands located within 12 Forest Districts (Fig. 1) being the main source of oak wood. The stands from which wood samples were taken for analysis are situated in seven Natural-Forest Regions: the Baltic Region (I) represented by two Forest Districts (Wejherowo, Świerczyna), Mazursko--Podlaska Region (II) – by one Forest District (Czarna Białostocka), Wielkopolsko--Pomorska Region (III) – by two Forest Districts (Krotoszyn, Milicz), Mazowiecko--Podlaska Region (IV) – by three Forest Districts (Jabłonna, Chełm, Mircze), Silesian Region (V) – by three Forest Districts (Wołów, Miękinia, Henryków), Małopolska Region (VI) – by two Forest Districts (Rogów, Tomaszów) and Carpathian Region (VIII) – by one Forest District (Kańczuga). Two trees with average diameter belonging

Fig. 1. Location of Forest Districts from where wood was col-
lected for measuring the content of heavy metals in the
stems of healthy and dying oak trees

Rys. 1. Lokalizacja nadleśnictw, w których pozyskano drewno do
badania zawartości metali ciężkich w pniach dębów zdro-
wych i zamierających

to the 1st and 2nd Kraft biosocial class were selected. They featured a straight stem and
absence of visible signs of diseases and injuries (cankers, cracks, fruiting bodies of
fungi, signs of insect occurrence). The trees represented two vitality categories accord-
ing to the classification by Roloff [1989] and Dmyterko [1998]: vital trees (0 or 0/1
degree) and damaged trees (3 or 2/3 degree). Oak wood samples were cut out of the
stem butt-end of trees aged 60-155 (Table 1). The trees were felled in the second half of
the growing season. The oak wood samples cut out of the outer heartwood were used for
testing. The samples contained ca 8-15 annual rings. The wood with an 8.5 per cent
moisture content was broken up in the Retsch grinder Ultra Centrifugal Mill ZM 1 into
sawdust. After sorting, the fraction which went through a 1 mm mash sieve and stopped
on a 0.5 mm sieve was used for testing. The wood was burnt at a temperature of 480°C
and mineralized with concentrated hydrochloric acid diluted with redistilled water at a
ratio 1:1.

Concentrations of heavy metals (Al, Cd, Cr, Cu, Fe, Mn, Pb) were marked using the
ICP-AES method in the Analytical Centre of the Warsaw University of Life Sciences –
SGGW.

A statistical analysis of the findings was carried out using STATISTICA 7.1. The
Mann-Whitney test was used to verify zero hypotheses.

Table 1. Characterisation of oak stands used for examining the relationships between healthiness of oak trees and heavy metal content in their wood

Tabela 1. Charakterystyka drzewostanów dębowych, w których pozyskano drewno do badań zależności między stanem zdrowotnym dębów a zawartością metali ciężkich w ich drewnie

Forest District Nadleśnictwo	Compartment or subcompartment Oddział, pododdział	Forest habitat type Siedliskowy typ lasu	Mean DBH $D_{śr}$ cm	Age, years Wiek, lata
Chełm	226d	Lśw	47.7	146
Czarna Białostocka	14b[a], 5a[b]	Lśw	36.0	105[a], 84[b]
Henryków	364b	Lśw	52.0	118
Jabłonna	146c	Lśw	28.2	60
Kańczuga	84a	LWyż	36.4	78
Krotoszyn	296a	Lśw	42.9	147
Miękinia	93d	LMśw	37.4	112
Milicz	49a	Lśw	49.4	123
Mircze	366a	Lśw	50.6	108
Świerczyna	20a	Lśw	33.7	110
Wejherowo	59c	LMśw	36.0	155
Wołów	336a	Lł	41.4	117

[a]Healthy tree. [b]Damaged tree. LMśw – fresh mixed broadleaved forest, LWyż – upland broadleaved forest, Lśw – fresh broadleaved forest, Lł – riparian forest.
[a]Drzewo zdrowe. [b]Drzewo zamierające.

RESULTS AND DISCUSSION

Mineral substances (ash)

Analysis of the content of mineral substances (ash) in the oak wood of healthy trees (0.1-0.9%) and dying trees (0.1-0.7%) showed that the ash content was within similar value ranges (Table 2). The mean ash content in the outer heartwood of healthy and dying oaks was at the same level equalling 0.3%. Literature data show that the ash content in oak wood is at the level of 0.2-0.7% [Prosiński 1984]. The ash content in sapwood is higher than in heartwood and its content is 0.5 and 0.3%, respectively [Fengel and Wegener 1989]. However, some non-European oak species (e.g. *Quercus douglasii*, *Q. stellata*) feature a significantly higher ash content reaching 1.4% [Rowell et al. 2005].

At five sites, the ash content in the wood of healthy oaks was higher than in the wood of dying trees. At three sites (Henryków, Wejherowo, Wołów), these values were at the same level. The lowest ash content was detected in the wood of dying oaks from Mircze, Kańczuga and Wejherowo and in the wood of healthy oaks from Krotoszyn and

Table 2. Ash content in the wood of healthy and dying oak trees from different regions of Po-
land, %

Tabela 2. Zawartość popiołu w drewnie dębów zdrowych i zamierających pochodzących z róż-
nych regionów Polski, %

Forest District Nadleśnictwo	Healthy tree Drzewo zdrowe	Damaged tree Drzewo zamierające
Chełm	0.4	0.7
Czarna Białostocka	0.4	0.3
Henryków	0.3	0.3
Jabłonna	0.9	0.3
Kańczuga	0.3	0.1
Krotoszyn	0.1	0.6
Miękinia	0.3	0.4
Milicz	0.2	0.5
Mircze	0.3	0.1
Świerczyna	0.6	0.4
Wejherowo	0.1	0.1
Wołów	0.3	0.3
Mean – Średnia	0.3	0.3

Wejherowo. The highest ash content was detected in the group of healthy trees from
Jabłonna (0.9%) and Świerczyna (0.6%), which can be linked to a relatively young age
of these two stands: ca 60 and 110 years, respectively. However, in the group of dying
trees, the highest ash content was detected at the Chełm (0.7%) and Krotoszyn (0.6%)
sites with the oldest oak stands whose dieback was a long-lasting process (Table 2).

The examined wood samples came from the stands representing four forest habitat
types (Table 1). The extreme ash content values were detected in the wood of both,
healthy and dying trees from the stands representing the fresh broadleaved forest habitat
type (Lśw).

The pH value of the tested wood of both, healthy and dying oaks was slightly acidic
and ranged from 3.40 to 4.26 [Piętka et al. 2005]. There are no significant differences in
wood between healthy and dying trees, however, in eight cases, the pH value of wood
in dying trees was slightly lower than in healthy trees. At six, out of eight, sites where
the wood pH was lower in dying trees, the ash content was lower (or at the same level)
in the wood of dying oaks compared to healthy ones.

Heavy metals

The data concerning the content of heavy metals show that the wood of healthy oaks
contained 33 to 192 mg/kg of dry mass of all examined elements (Table 3). Although
the lower value was almost six-fold less than the upper value, these values should be
considered relatively small.

Table 3. Total content of the analysed heavy metals in the wood of healthy and dying oak trees
         from different regions of Poland, mg/kg of dry mass
Tabela 3. Sumaryczna zawartość badanych metali ciężkich w drewnie dębów zdrowych i zamiera-
         jących pochodzących z różnych regionów Polski, mg/kg suchej masy

Forest District Nadleśnictwo	Healthy tree Drzewo zdrowe	Damaged tree Drzewo zamierające
Chełm	192.29	201.54
Czarna Białostocka	136.72	127.06
Henryków	123.54	95.79
Jabłonna	110.94	98.29
Kańczuga	88.15	78.23
Krotoszyn	83.24	98.88
Miękinia	88.45	87.42
Milicz	32.64	79.45
Mircze	59.02	41.26
Świerczyna	62.54	233.60
Wejherowo	98.37	80.38
Wołów	78.13	71.91

No impact of the geographic location of stands on heavy metal concentrations in oak wood was observed. The lowest content of heavy metals was detected in the oak wood from Milicz (Wielkopolska) – 33 mg/kg of dry mass, while the highest – in Chełm – 192 mg/kg and Czarna Białostocka – 136 mg/kg of dry mass, that is from the stands representing Forest Districts located in the eastern region of Poland (Table 3). No significant differences were found between concentrations of heavy metals in the wood from the remaining Forest Districts, as for example the wood from Henryków located in the Lower Silesia Region contained 123 mg/kg of dry mass, and the wood from Wejherowo (the Baltic Region) – 98 mg/kg of dry mass (Table 3).

The wood of dying oaks and the wood of healthy oaks had a similar content of heavy metals. The total content of examined heavy metals in the oak wood samples ranged from 41 mg/kg of dry mass (Mircze) to 233 mg/kg of dry mass (Świerczyna; Table 3), except for the wood from Świerczyna, where the content of examined metals in the dying tree was four-fold higher compared to the wood of the healthy tree (Table 3). This result was impacted by the exceptionally high level of manganese (153 mg/kg of dry mass), i.e. several or even a dozen or so times higher than in the remaining wood samples. The wood from the Świerczyna Forest District is represented by the pedunculate oak (*Quercus petraea*), a species which, in the opinions of some authors, features higher susceptibility to heavy metal concentrations in the environment than other oak species [Opydo et al. 2002]. Such differences are and were observed between species from the genus poplar (*Populus*) [Łukaszewski et al. 1993]. It is likely that such a high concentration level of manganese in the pedunculate oak wood was the result of a high content of this easily assimilative element in soil. On the other hand, the wood of healthy pedunculate oak from Świerczyna contained a several-fold lower content of this element.

As the total content of heavy metals in the oak wood samples was relatively low, the cation concentrations were also low, yet these values were not equal (Table 4). The content of Cd, Pb and Cd in the examined material was at the lowest level (1 mg/kg of dry mass, on average). The Cd content in wood samples fell within the range 0.02-0.21 mg/kg, Pb – 0.17-0.86 mg/kg and Cr – 0.16-0.86 mg/kg. In spite of voluminous literature on the occurrence and negative effects of these metals, no evidence has been found so far that might confirm that the examined elements are indispensable for normal plant development [Kabata-Pendias and Pendias 1999]. The values for Cd and Pb concentrations did not exceed the level accepted as natural for the wood of oak species [Queirolo

Table 4. Content of heavy metals in the wood of healthy and dying oak trees, mg/kg of dry mass
Tabela 4. Zawartość metali ciężkich w drewnie dębów zdrowych i zamierających, mg/kg suchej masy

Tree health grade Stopień zdrowotności drzewa	Heavy metals – Metale ciężkie mg/kg				
	minimum minimum	maximum maksimum	mean średnia	SD	Vx
Al					
Healthy – Zdrowe	6.56	124.02	55.91	30.53	54.61
Damaged – Zamierające	7.96	127.23	57.95	25.82	44.55
Cr					
Healthy – Zdrowe	0.16	0.56	0.27	0.17	41.40
Damaged – Zamierające	0.17	0.86	0.31	0.17	57.16
Cu					
Healthy – Zdrowe	1.17	16.02	2.22	2.96	133.44
Damaged – Zamierające	1.18	2.14	1.69	0.29	17.25
Fe					
Healthy – Zdrowe	4.93	24.82	11.70	5.65	48.31
Damaged – Zamierające	1.14	42.82	12.22	8.25	67.49
Mn					
Healthy – Zdrowe	3.32	114.22	25.67	24.06	93.74
Damaged – Zamierające	2.96	153.72	34.37	43.26	125.87
Pb					
Healthy – Zdrowe	0.20	0.51	0.37	0.12	33.06
Damaged – Zamierające	0.17	0.86	0.39	0.17	43.58
Cd					
Healthy – Zdrowe	0.02	0.11	0.03	0.02	67.21
Damaged – Zamierające	0.02	0.21	0.04	0.04	91.28

SD – odchylenie standardowe, Vx – współczynnik zmienności.
SD – standard deviation, Vx – variability coefficient.

et al. 1990, Opydo and Opydo 1996, Opydo et al. 2002, 2005]. Also, the content of Cd and Pb in the wood of oak seedlings grown on control plots does not exceed 1 mg/kg of dry mass [Greszta 1982].

Although no statistically significant difference in the Cr content was found in the wood of healthy and dying oaks, attention should be drawn to the fact that the concentration of this element was higher in the wood of dying trees than in the wood of healthy trees at all sites except for Wejherowo. If the content of Cr in the healthy wood is assumed as 100%, the content of this element in the wood of dying oaks oscillated from several per cent (Miękinia, Świerczyna, Wołów) to 150 per cent more (Milicz). Relevant literature lacks data on the natural level of Cr in oak wood. On the basis of the findings, it can be suggested that the natural level of this element does not exceed 1 mg/kg of dry mass. The concentration of this microelement in the wood of other tree species, e.g. European beech (*Fagus sylvatica*) ranges from trace quantities up to 0.05 ppm [Kaźmierczakowa et al. 1984]. In edible crops, the Cr concentration has a quite broad range 0.02-1 ppm. It is generally assumed that the concentration of Cr > 2 ppm for extremely susceptible plants and of Cr < 20 ppm for moderately resistant plants is damaging [Kabata-Pendias and Pendias 1999].

The content of copper cations in the tested oak wood ranged from several to a dozen or so mg/kg of dry mass which is a natural level of this element in soil [Kabata-Pendias and Pendias 1999]. Both, the deficiency and excess of copper, which is an indispensable element for the normal development and growth of plants, lead to disturbances in their various physiological processes. The range of Cu concentrations in the wood of healthy oaks (1.17-16.02 mg/kg) was greater than in the wood of dying oaks (1.18-2.14 mg/kg). However, in all wood samples, the content of copper was below 2.5 mg/kg except for the sample from the Wołów Forest District where it was markedly higher (16.02 mg/kg). The obtained results do not confirm the results of the studies by Opydo and Opydo [1996] who observed significantly higher Cu concentrations in the wood of dying oaks compared to healthy ones.

The content of iron in the wood of healthy oaks was 4.93-24.82 mg/kg, while in the wood of dying oaks – 1.14-42.82 mg/kg (Table 4). At five sites (Chełm, Czarna Białostocka, Krotoszyn, Milicz, Mircze), the Fe content was higher in the wood of dying oaks than in the wood of healthy oaks and the differences were between 20% (Mircze) and over 100% (Milicz). At the remaining sites, the content of Fe was at a similar level or slightly higher in the wood of healthy trees compared to dying ones.

The contents of aluminium and manganese in the oak wood samples were highest (Table 4). The content of aluminium was 6.56-124.02 mg/kg of dry mass in the wood of healthy trees, while 7.96-127.23 mg/kg of dry mass in the wood of dying trees (Table 4). At seven sites, the Al content was higher in the wood of dying trees compared to healthy ones, while for the majority of cases the differences were minor. The exception was the wood from Milicz and Świerczyna where Al concentrations were five-fold and three-fold higher in the wood of dying trees compared to healthy ones, respectively.

Aluminium is a common element in plants usually not exceeding the concentration range of X0-X00 ppm, however, its content significantly differs depending on plant properties and soil conditions. A relatively high Al content in wood should be related to the fact that it is one of the most widespread elements in the lithosphere (7.45%) and it was man who has caused its amplified cycling in nature. Lévy et al. [1996] pointed to a marked increase of Al content in pedunculate oak wood from a dozen or so to several

dozen mg/kg of dry mass during several decades in the second half of the 20th century. Elevated soil pH causes leaching of manganese, potash and calcium compounds and release of Al^{+3} ions which have a toxic effect on the root system of trees leading to a dieback of fine roots and micorrhizae [Prusinkiewicz and Pokojska 1989].

The content of manganese was 3.32-114.22 mg/kg of dry mass in the wood of healthy oaks and 2.96-153.72 mg/kg of dry mass in the wood of dying oaks (Table 4). Out of four sites, where the Mn concentration in the wood of dying oaks was higher than in the wood of healthy oaks, it was the Świerczyna site where differences between the examined trees were largest. At this site, the content of manganese in the wood of dying oaks (152.83 mg/kg) was over six-fold higher than in the wood of healthy oaks (24.08 mg/kg). In the group of sites at which Mn concentrations in the wood of healthy oaks were higher than in the wood of dying oaks, the greatest difference (four-fold) was detected at the Krotoszyn site. Manganese, whose concentrations in oak wood during these studies were highest, plays important metabolic functions in plants, and its content can oscillate in a broad range, depending on the species, plant part, age, storage of as-similative manganese in soil and other factors [Kabata-Pendias and Pendias 1999].

The comparison of significance of differences in the concentration of heavy metals in the wood of healthy and dying oaks was performed. The values of this parameter for most elements had no normal distribution (Shapiro-Wilk test). Therefore, decisions were taken to use a Mann-Whitney non-parametric test ($\alpha = 0.05$). No ground was found for each of the examined elements to reject a zero hypothesis of the lack of dif-ferences in element concentrations in the groups of healthy and dying oak trees.

CONCLUSIONS

The findings allow the conclusion that no significant differences in the content of heavy metals in the outer heartwood were found between oaks which were affected by the process of dieback irrespective of the duration of this phenomenon (from few to several dozen years) and healthy oaks growing under the same environmental condi-tions, i.e. in the same stands. In the case of some heavy metals, a rising tendency was observed pointing to their higher levels in the wood of dying than in the wood of healthy trees. Out of the 12 examined sites, the concentrations of Cr at eleven sites and of Al and Pb at seven sites were found higher in the wood of dying oaks compared to healthy ones. The content of Mn and Fe was higher in the wood of healthy oaks at the majority of sites.

No impact of the geographic location of stands on heavy metal concentrations in oak wood was observed. On the basis of the examined chromium content, it can be con-cluded that the natural level of this element in the heartwood of oaks does not exceed 1 mg/kg of dry mass.

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ZAWARTOŚĆ METALI CIĘŻKICH W DREWNIE DĘBÓW (*QUERCUS ROBUR* L., *Q. PETRAEA* (MATT.) LIEBL.) ZDROWYCH I ZAMIERAJĄCYCH

Streszczenie. Przeprowadzono badania ogólnej zawartości substancji mineralnych (popiołu) i metali ciężkich (Al, Cd, Cr, Cu, Fe, Mn, Pb) w drewnie dębów zdrowych i zamierających z różnych rejonów Polski. Materiał do badań pochodził z 12 nadleśnictwach reprezentujących główne bazy surowca tego gatunku w Polsce. Z każdego drzewostanu, w którym obserwowano w przeszłości i obecnie objawy zamierania oraz nadmiernego wydzielania się drzew, wycięto w tym samym wieku dwa drzewa: zdrowe i zamierające. Analizowano materiał pozyskany z twardzieli zewnętrznej odziomkowej części pni dębów w wieku 60-155 lat. Zawartość substancji mineralnych w drewnie dębów zdrowych i zamierających była bardzo podobna. Nie stwierdzono statystycznie istotnej różnicy dla każdego z badanych pierwiastków w ich zawartości w drewnie drzew zdrowych i zamierających.

Słowa kluczowe: metale ciężkie, popiół, dąb szypułkowy, dąb bezszypułkowy, drewno dębowe, Polska, stan zdrowotny, witalność drzew, zamieranie lasu