EFFECTS OF OPEN-CAST SULPHUR MINING ON SEDIMENT TRANSFERS AND TOXIFICATION OF RIPARIAN FORESTS

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ABSTRACT. The spoil heaps of the sulphur mines of the Negoiul Românesc volcanic cone are intensely reworked by mass movement processes which not only lead to severe sedimentation in downstream rivers, but also to the toxification of riparian Norway spruce (*Picea abies* (L.) Karst.) forests. Along the lateral borders of the Dumitrelul retention basin, recent sedimentation has covered *P. abies* trees with up to 160 cm of toxic, sulphur-rich material originating from the spoil heaps deposits. This study documents the deposition of toxic sediments and illustrates the effects that these sulphur-rich sediments have on tree growth. The results show that trees affected by sedimentation reacted with severe growth suppression and the formation of tangential rows of traumatic resin ducts, and that the intensity and persistence of growth anomalies in trees are positively correlated with local depths and granulometry of sediments.

Key words: Norway spruce forest, tree-rings, growth anomalies, Călimani Massif (Romania)

Introduction

Sediment transfers in poorly consolidated spoil heaps deposits may represent a serious threat for the environment and people living downstream of abandoned mining sites. The Negoiul Românesc volcanic cone (Călimani Massif, Romania, 47° 07′ 00″ N; 25° 13′ 38″ E; 1895 m a.s.l.) is such a case, and toxic debris flows have recently caused intense sedimentation of downstream river valleys and toxification of riparian Norway spruce (*Picea abies* (L.) Karst.) forests located roughly between 1700 and 1300 m a.s.l., with substantial associated risks for the villages downstream of the site and the ecosystems of the riparian forests.

The transfer of toxic sediments from active and abandoned mines and the contamination of aquatic environments as a consequence of deep metal mining for Pb, Zn, Cu, Cd and Fe is of widespread international concern, as pollution resulting from metal mining activities can result in significant environmental and ecological degradation and can pose serious risks to human health through contamination of food and drinking water (Byrne *et al*. 2012). Herr and Gray (1996), for instance, assessed seasonal variations of metal contamination of riverine sediments below a Cu and S mine in southeast Ireland and point to the major role of hydrological factors in metal accumulation of highly energetic river systems. Byrne *et al*. (2013) confirm the crucial role of extreme events (in the sense of stormflows, floods or debris flows) on hydrochemistry of contaminated drainage from (abandoned) mines in Wales, such that metal pollution from mining can contaminate river systems to a degree at which it may significantly hinder the achievement of European Union Water Framework Directive objectives (Lynch *et al*. 2014).

The burial of trees and related growth effects have been the subject of a handful of papers as well, but none has assessed the combined effect of increased anaerobicity (as a result of sedimentation) and toxification (sulphur uptake) so far. Burial of plants, beyond their threshold level of tolerance, has been found to result in abiotic stresses which may eventually filter intolerant species out of the com-

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munity (Maun 1998; Dech 2004; Dech and Maun 2006). Growth reactions of plants to burial have been documented in sand dune environments with a focus on burial tolerance and adaptive traits (e.g. adventitious root formation, plastic resource allocation to biomass, rapid vertical elongation of shoots) in saplings of woody plants (e.g. Singh and Rathod 2002; Shi *et al*. 2004; Liu *et al*. 2007; Zhao *et al*. 2007). In contrast to the large body of experimental studies focusing on burial effects in saplings, work on mature plants remains scarce. Pioneering studies in the field demonstrated that stem burial is likely to result in pronounced narrowing of radial growth as the most frequent and obvious reaction to burial (e.g. Hupp 1988; Friedman *et al*. 2005). Strunk (1991) reported that mature and old-growth Norway spruce (*P. abies* (L.) Karst.) trees are capable of surviving multiple burial events of up to 1.7 m in dolomitic depositional environments. Kogelnig-Mayer *et al*. (2013) recently documented the effects of abrupt stem burial on growth in mature European larch (*Larix decidua* Mill.) trees by debris flows. They found that abrupt burial causes massive suppression of radial growth compared with pre-event conditions such that the affected trees were unable to resume pre-burial growth rates even after 25 years, irrespective of sampling height along the stem axis.

Based on the above considerations, this study aims to date recent sedimentation in a reservoir located underneath an abandoned sulphur mine area, and analyse growth reactions of *P. abies* trees to burial and toxification by sediments so as to determine temporal and burial depth thresholds that trees can support in such environments.

Study site

The Călimani massif (Fig. 1a) is formed by the highest stratovolcano of the Carpathian volcanic chain (Pietrosul peak, 2100 m a.s.l.). The massif was built by effusive, explosive, extrusive and intrusive processes, followed by a caldera collapse sometime between 11.9 and 6.7 Ma (Pécskay *et al.*

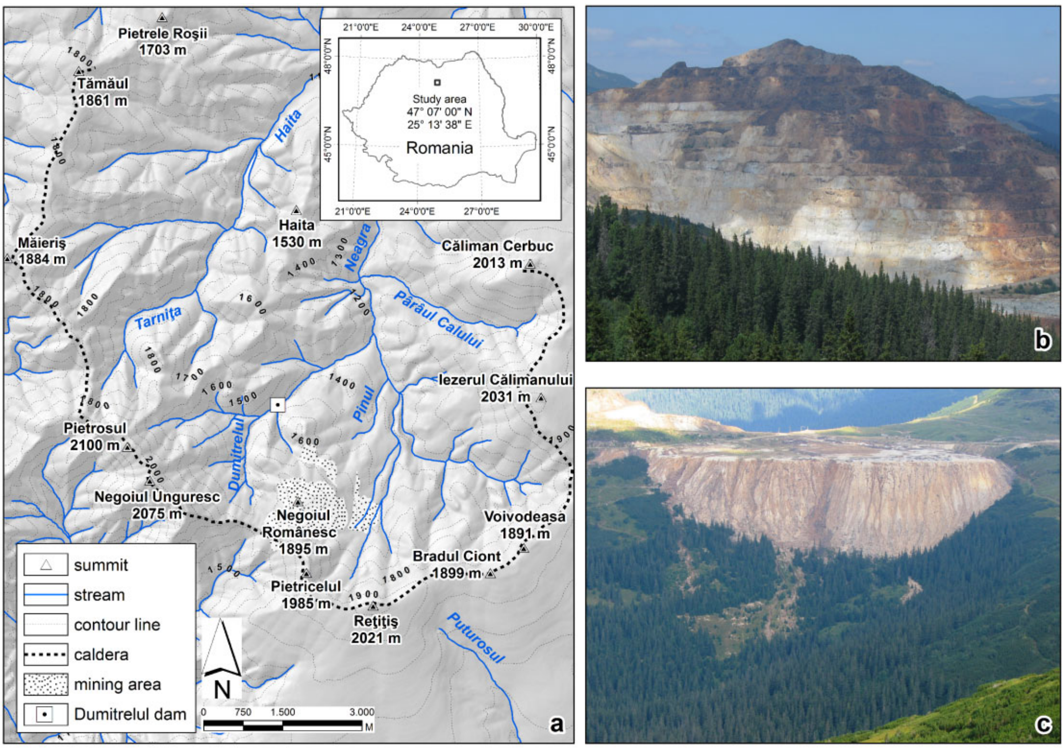


Fig. 1. (a) Map location of the study area. The map symbols indicate: (1) main peaks; (2) drainage network; (3) opencast sulphur mining area; (4) Dumitrelul sediment retention basin location. (b) The opencast sulphur mine on the Negoiul Românesc stratocone (view facing east). (c) Dumitrelul spoil heap (view facing west), the main source area of debris flows which then propagate downstream for 3 km to be deposited in the Dumitrelul reservoir (photos of the figure 1b and 1c by O. Pop, 2008).

1995a, 1995b; Szakács and Seghedi 2000; Seghedi *et al*. 2005; Szakács and Krézsek 2006). A lava dome has subsequently formed along the southern rim of the caldera, where fumerolian and solfatarian activity has then created sulphur-rich volcanic ore deposits at what is nowadays known as the Negoiul Românesc dome.

An opencast sulphur mine (Fig. 1b) was operational at Negoiul Românesc between 1966 and 1997, resulting in spoil heaps piled up on the steep slopes next to the pit. Sulphur ore exploitation was most intense between 1976 and 1986 (Kern *et al*. 2009), and resulted, amongst other things, in the progressive formation of the Dumitrelul spoil heap (Fig. 1c). After the cessation of mining activities, it reached a volume of *c*. 6.5 million m^3. The platform of the Dumitrelul spoil heap covers 110 000 m^2 and its talus has a surface of >25 000 m^2. The material transported by torrential processes originates mainly from this talus surface and has a granulometry ranging from silt to blocks of up to 5 m in diameter. Mobilization typically occurs during heavy rainfalls when water accumulates on the spoil heap platform to flow down the talus in ever increasing gullies.

Neither rehabilitation works to stabilize the deposits, nor erosion control measures have been undertaken after the closure of the mine in 1997. As a consequence of the morphological instability, acid mine drainage and the lack of a soil layer on the spoil heaps, natural reforestation did not take place and thus prevented the consolidation of the material in the source area (Surdeanu *et al*. 2011), which in turn favoured the release of debris flows and floods during storms.

The Dumitrelul retention dam (Fig. 2) is located 3 km downstream of the spoil heap at 1322 m a.s.l. and was built between 1980 and 1982, when excessive sedimentation first occurred in the Dumitrelul River downstream of the spoil heap area. The reservoir was designed to retain 30 000 m^3, but was almost completely full in 2007 and after a series of debris flows (Pop *et al*. 2009). Severe sedimentation continues to occur in the area, even more than 15 years after the discontinuation of sulphur mining, and increasingly affects the riparian forest composed of dense stands of Norway spruce (*P. abies* (L.) Karst.) trees. The reservoir was emptied in 2009, and trees located inside the reservoir (buried by up to 3 m of sediment) were removed. Until 2009, trees growing at the lateral borders of the reservoir were buried by up to 1.6 m of sediment, but left at the site after excavation works.

Fig. 2. Dumitrelul sediment retention reservoir, before sediment excavation in 2007 (a) and after excavation works carried out in 2009 (b) (figures from Pop *et al*. 2013).

Material and methods

Topographic field measurements

During the field campaigns in summer 2007 and spring 2010, topographic surveys were carried out in the basin to quantify the infilling of the reservoir. All measurements were carried out using a Leica 407 total station. These changes in infilling then served for the calculation of average sedimentation rates. In addition, the total station was used to localize the sites selected for granulometric analyses and tree-ring analyses.

Granulometric analysis of reservoir's sediments

Granulometric analyses were carried out within sites located in the vicinity of buried trees, namely in the lower, central and upper portions of the reservoir (Fig. 3). Samples were taken from each sediment layer in the profile and sieved at the

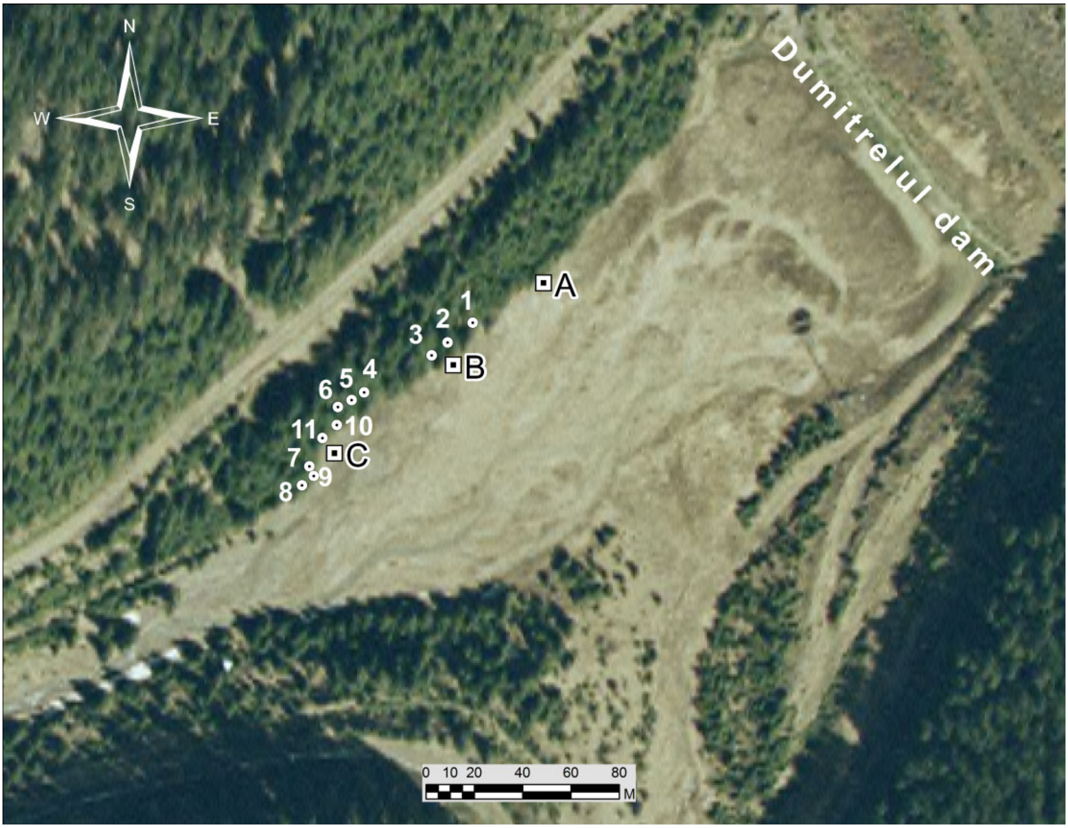


Fig. 3. Sampling sites within the Dumitrelul reservoir: rectangles indicate locations of granulometric profiles whereas circles indicate positions of sampled trees. (figure from Pop *et al.* 2013, modified)

laboratory with Retsch sieves to obtain sediment volumes by particle size classes (Udden-Wentworth scale).

Tree sampling design

Based on a first reconnaissance at the site and the univocal detection of massive and widespread anatomical reactions in trees affected by toxification (unpublished data), trees were systematically sampled in spring and autumn 2010 with a focus on living trees growing along the left border of the reservoir. We sampled 11 buried trees (i.e. absence of roots at the stem base, burial of lowermost branches, signs of reduced vitality such as brown crowns or dying bark) (Fig. 3). Tree positions were determined with a Leica 407 total station. We then excavated the trees down to the root collar level, and took samples every 10 cm intervals on the stems and up to a height of 50 cm measured from the sediment level reached in 2009 and before the excavation works. Samples gathered from damaged trees included 252 increment cores (from eight trees) and 48 cross-sections (from three trees). Sampling direction was systematically recorded on the cores as illustrated in Fig. 4. In a next step, increment cores were taken from 20 undisturbed *P. abies* trees growing next to the reservoir and outside the reach of toxic sediments (two cores per tree extracted perpendicular to the slope, 40 cores in total), so as to obtain a local reference chronology built of trees that were not affected by sulphur-rich sediments. Reference trees were selected upslope, at several tens of meters from the reservoir.

Tree-ring analysis

In the laboratory, increment cores and cross-sections were prepared for analysis using standard dendrochronological procedures, including air-

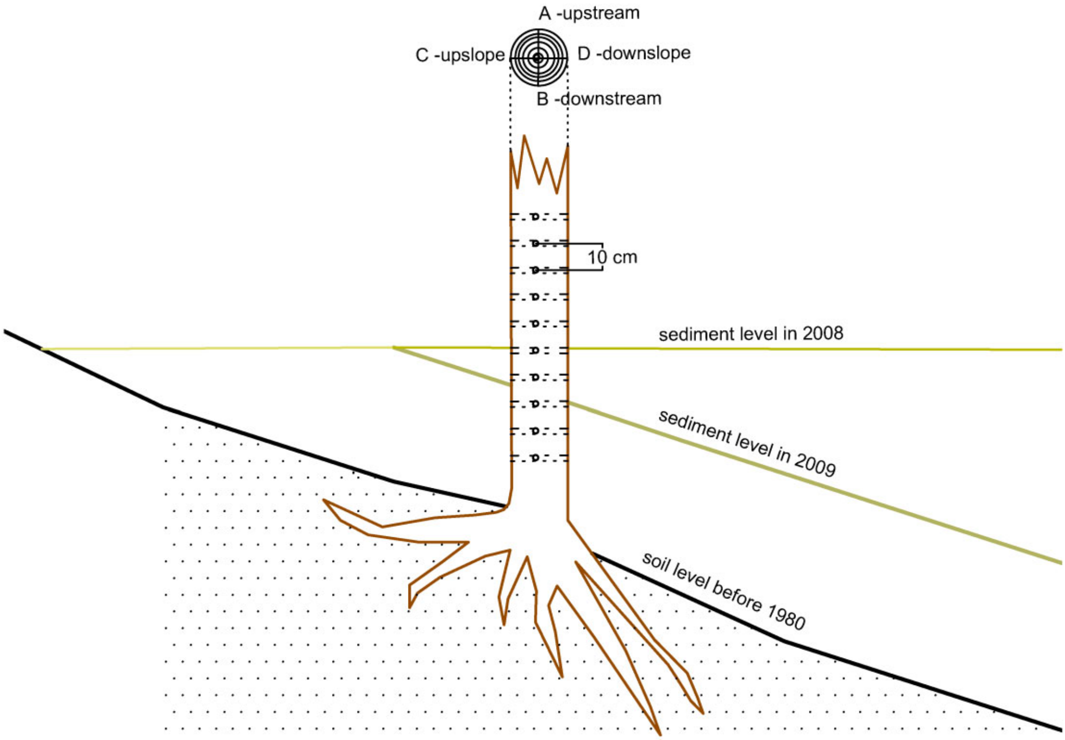


Fig. 4. Schematic illustration of sedimentation of *Picea abies* (L.) Karst. trees with indication of sampling directions around and along the stem (figure from Pop *et al.* 2013, modified).

drying, sanding and tree-ring counting (Stoffel and Bollschweiler 2008; Stoffel and Corona 2014). Ring widths were then measured on all cross-sections and cores using a digital LINTAB 5 system (a positioning table connected to a Leica stereomicroscope and computer with TSAP-Win™ Professional software; Rinntech, http://www.rinntech.de/content/view/17/48/lang,english/index.html, 14 Oct., 2014).

The reference chronology obtained from 20 undisturbed trees was built following the same procedures and served the cross-dating of affected trees; it helped the detection of missing rings and the distinction of geomorphic and/or toxic disturbances from climatic fluctuations (Cook and Kairiukstis 1990).

One of the most typical reactions in trees after burial is *growth suppression* (*GS*) (Kogelnig-Mayer *et al*. 2013). After sedimentation, roots suffer from reduced activity, but the mechanical effects caused by the enormous weight of debris have been reported to affect tree growth processes as well. The pressure on the cambium exerted by the bark and phloem impedes the cell division and leads to a reduced number of cells with narrower lumen (Kny 1877), resulting in a reduction of ring width to 25% of the original width (Rubner 1910). The focus of this study consequently is also on GS, and a reduction by more than 100% compared with the previous ring width is considered an appropriate indicator of severe burial. GS was recorded if ring widths were more than 100% smaller than previous growth for at least two consecutive rings. Calendar dates of GS sequences were identified on all cross-sections of the trees sampled. Initiation and extension of GS was observed axially in all samples of an individual tree before we compared the reactions of individual trees with those found in the tree-ring series of other trees.

In addition to GS, we also focused on the occurrence of tangential rows of *traumatic resin ducts* (*TRDs*) (Stoffel 2008). TRDs generally form in the wood and bark of spruce (*Picea* species) and typically consist of closely aligned, thin-walled, long-lived secretory epithelial cells (Nagy *et al*. 2000). The formation of TRDs in the developing secondary xylem has been reported after insect attack, fungal elicitation or drought stress (Fahn *et al.* 1979; Langenheim 2003; McKay *et al.* 2003; Plant 2003; Hudgins *et al.* 2004; Krekling *et al.* 2004). Abundant occurrence of TRDs has also been reported as a result of mechanical wounding by (hydro-) geomorphic processes, such as snow avalanches, rockfalls or debris flows (Stoffel *et al.* 2005; Bollschweiler *et al.* 2008; Stoffel and Hitz 2008; Schneuwly *et al.* 2009a, 2009b). It is assumed here that the presence of toxic sediments originating from the spoil heap area will cause the formation of TRDs as well. Reactions of the same nature occurring in the same tree over several years have been identified as a continuing response of the tree to the same initial disturbance (Stoffel and Corona 2014). TRDs were also analysed in the reference trees so as to separate years with widespread resin formation caused by insect attacks or droughts from severe TRD events induced by geomorphic processes (Stoffel 2008) or geochemical stress.

Results

Recent sedimentation rates in the Dumitrelul reservoir

The assessment of sedimentation rates in the Dumitrelul reservoir was based on repeat topographic surveys in the field between 2007 (Pop *et al.* 2009) and 2010. The reservoir was designed to retain 30 000 m^3 of sediment reworked from the spoil heap area in 1982, but almost reached the maximum infill level in summer 2007 (25 500 m^3; length 190 m; width 80 m; surface area 9260 m^2), thus yielding an average sedimentation rate of 1020 m^3 yr^{-1} for the 1982–2007 period. The reservoir was emptied in 2009 when roughly 26 800 m^3 of toxic sediments were removed.

Granulometric analysis

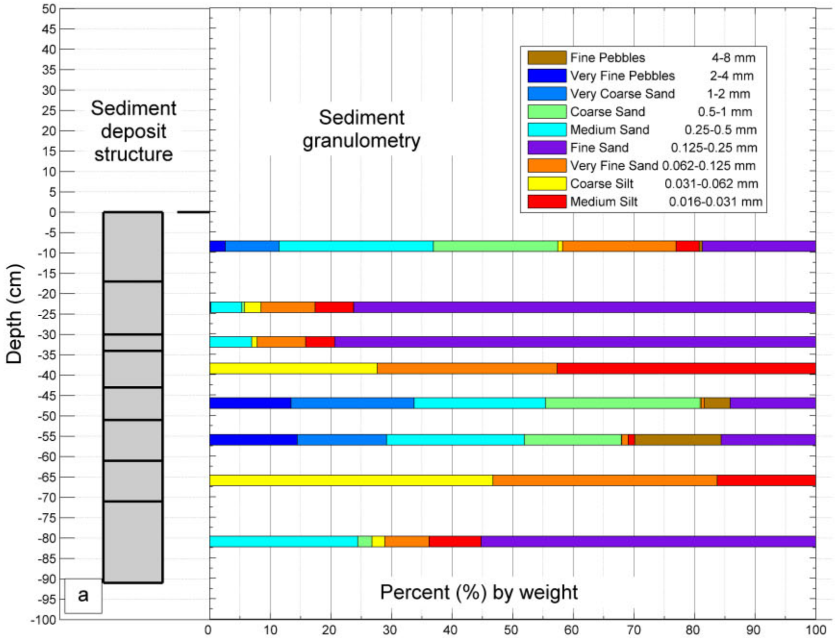
The distribution of particle size classes of each sediment layer as well as the location of the three sampling sites are given in Fig. 5a–c. In profile A (depth 92 cm; Fig. 5a) located in the lower sector of the reservoir, a succession of eight distinct sediment layers can be identified, each of them being constituted by a variable proportion of particle size classes. In general terms, fines (sand and silt) show a large distribution in each of the sediment layers analysed. In profile B (depth 50 cm; Fig. 5b), realized in the central sector of the reservoir, four sediment layers can be found, each of them having a variable proportion of size classes, but again with a clear predominance of sand and silt. Profile C, located in the upper sector of the reservoir (Fig. 5c), presents a different context with more undifferentiated layers and a granulometry dominated by pebbles.

Growth anomalies in trees: determination of severe sedimentation events

The germination dates of the trees sampled inside the reservoir ranged between 1965 (tree 1) and 1986 (tree 11; Table 1). The frequency of GS and TRD reactions are presented in Fig. 6. The first year with growth anomalies occurred in 1990 (tree 4), followed by reactions in 1992 in trees 6 and 10, and later on in tree 2. The years 1995 and 1996 seem to refer to a single severe sedimentation episode, as indicated by severe GS in six different trees. Similarly, the GS in 2000 and 2001 also refer

Table 1. Database of trees analysed and growth reactions.

Tree	Circumference (cm)	Burial depth (cm)	Number and type of samples	Germination
1	86	65	32 increment cores	1965
2	53	75	32 increment cores	1977
3	83	0	20 increment cores	1974
4	91	0	20 increment cores	1970
5	57	80	40 increment cores	1979
6	87	30	24 increment cores	1977
7	91	40	32 increment cores	1976
8	80	160	52 increment cores	1984
9	29	70	13 stem discs	1978
10	38	120	17 stem discs	1983
11	36	120	18 stem discs	1986



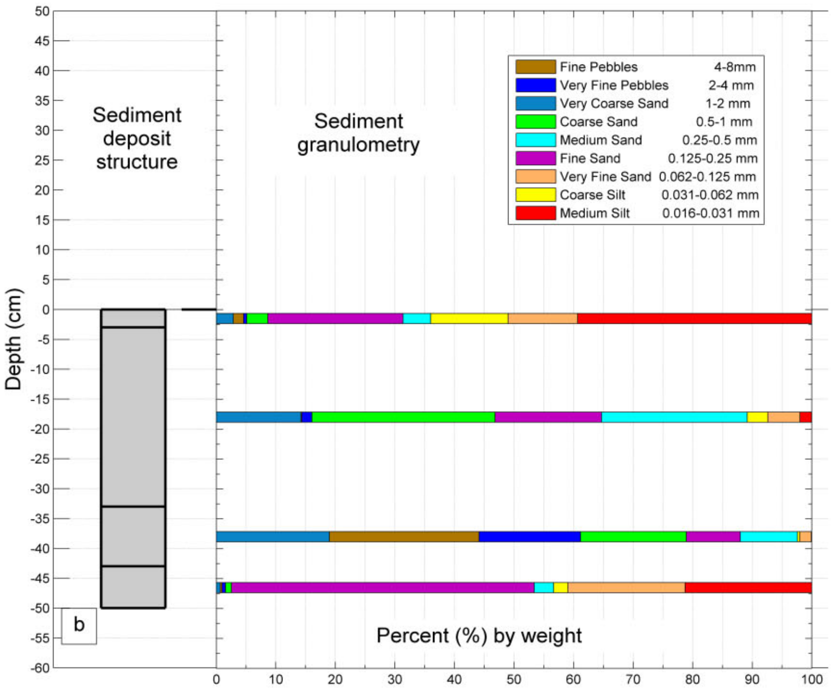


Fig. 5. (a–c) Granulometric characteristics of the sediment deposits.

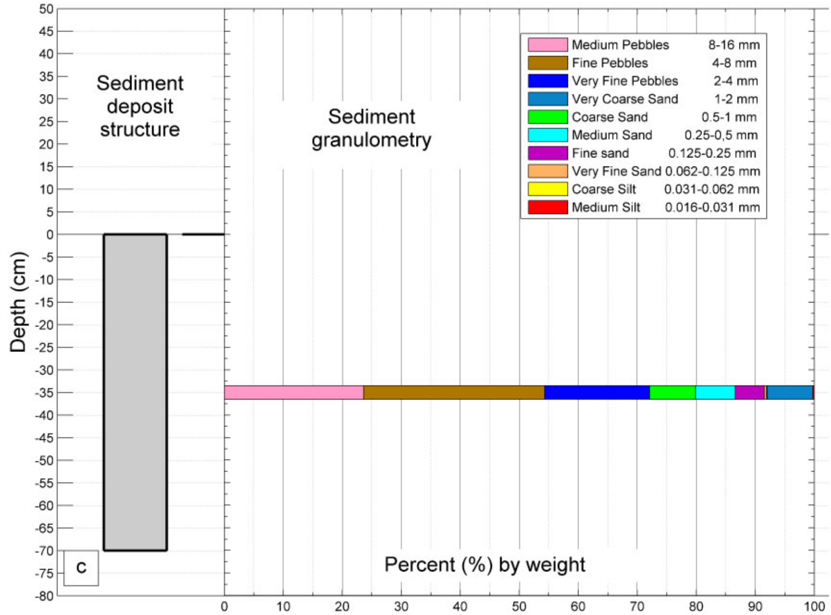


Fig. 5. Continued

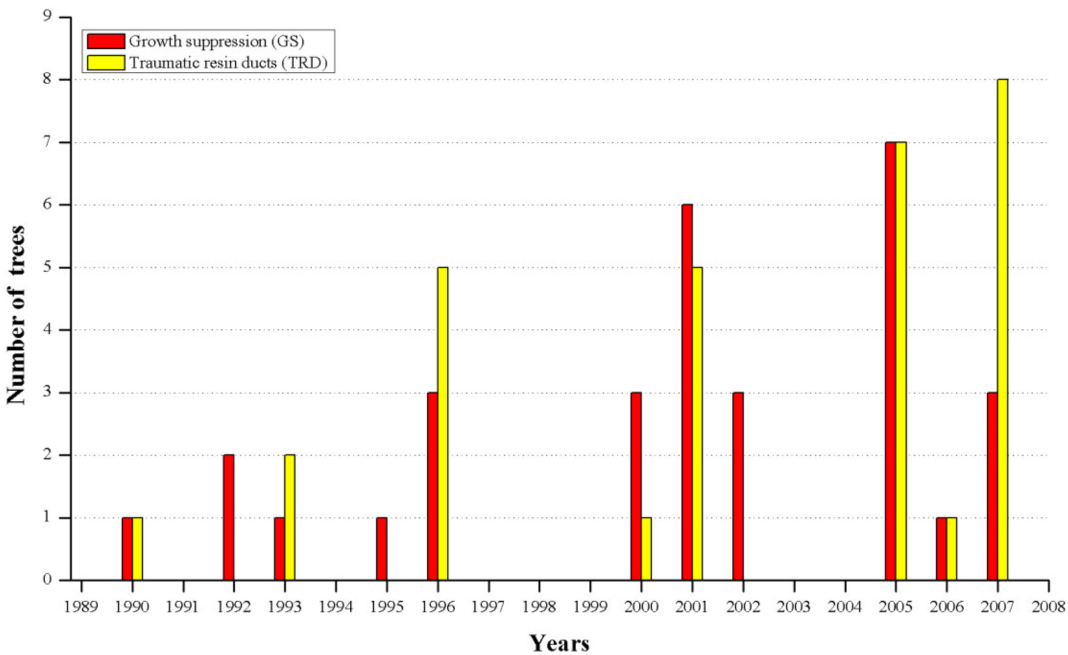


Fig. 6. Temporal frequency of GS and TRD occurrences in the growth-ring records of the trees sampled and for the period 1990–2007.

to a single sedimentation episode and are recorded in nine trees. The flows in 2005–2006 are represented by reactions in eight trees. The last sedimentation event occurred in 2007 and is recorded in all but one tree (10 trees). In addition to the GS, trees also formed TRDs as a reaction to stress. Interestingly, as shown in Fig. 6, the occurrence of TRDs is simultaneous of that of GS, which means that

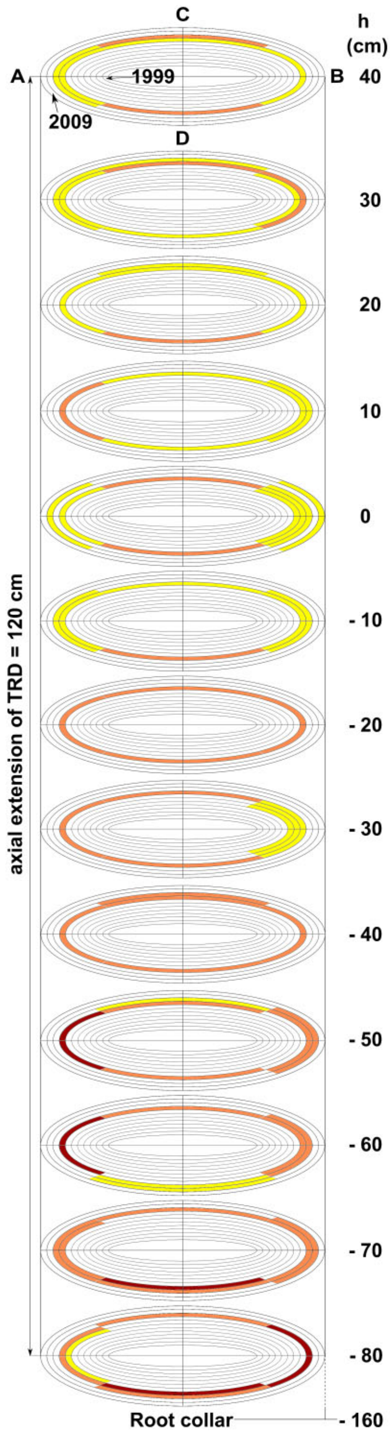


Fig. 7. Axial distribution of TRD in tree 8 following toxification by contaminated, sulphur-rich water. Colours indicate the TRD intensity in different years and at different sampling levels: red/dark grey – compact band of tangentially oriented resin ducts with almost no space between ducts; orange/medium grey – tangentially oriented ducts with several spaces between ducts; yellow/light grey – individual ducts, somehow aligned.

resin was produced as a result of stem base burial by toxic sediment, but not by the burial itself. We also observed that GS typically precedes (or occurs simultaneously to) TRD formation, or that at least TRD formation does not occur before the severe burial (e.g. trees 2, 6 and 10). At the same time, we realize that in tree 8 reactions are first in the form of TRD formation. In this case, shallow sedimentation, or the contamination of the water table with toxic waters, could have affected its vitality and that important sedimentation only occurred at a later stage.

Temporal and level occurrence of growth anomalies

With respect to the propagation of growth anomalies within and along the stem exhibit an axial distribution of reactions to up to 120 cm below the maximum sediment surface level reached in 2009, but also up to 50 cm above this level. In all buried trees GS sequences extend axially and over the entire segment investigated, and the abrupt GS persists for >5 yr on average. The suppression was largest after the massive burial event in 2007 when between one and three rings were missing at the stem base. TRDs are often being formed in the year of GS formation, but in some trees reactions start after a delay of one or more years. An example of the axial distribution of TRDs is presented in Fig. 7.

Discussion

In the study presented here, the infilling of the Dumitrelul reservoir has been documented using repeat topographic surveys, as well as sedimentation events as recorded in trees. In addition, this study addresses the impacts that the toxification of Norway spruce (*P. abies* (L.) Karst.) with sulphur-rich water has on its vitality. We clearly illustrate that the combination of topographic measurements, granulometric analysis and tree-ring records represent useful tools to reconstruct the history and impacts of sedimentation processes on vegetation in the Dumitrelul reservoir.

The most important reaction to sedimentation observed in this study are in the form of abrupt GS in the tree-ring records of the *P. abies* trees, a feature which could be observed in all trees sampled and which extended axially at all heights considered. Under the most extreme conditions (i.e. extreme sedimentation in 2007), rings were even missing completely in some of the cross-sections taken at the stem base. Our study confirms the findings of Kogelnig-Mayer *et al*. (2013) who previously described GS as the main growth anomaly in trees after stem burial.

In addition to GS, we also observed excessive amounts of tangential rows of TRDs following sedimentation, namely in the same years as GS formation. The occurrence of TRDs has so far been described as a consequence of mechanical wounding (Bollschweiler *et al*. 2008; Stoffel and Hitz 2008; Schneuwly *et al*. 2009a, 2009b) and/or the result of drought stress, but has not been observed in relation – at least not in significant numbers – in trees affected by burial (Kogelnig-Mayer *et al*. 2013). We therefore assume that the massive formation of TRDs is clearly reflective of the toxification event and the deposition of toxic sediments. This assumption is further confirmed by the absence of TRDs in the reference trees growing next to the Dumitrelul basin so that air pollution (Hättenschwiler *et al*. 1996; Muzica *et al*. 2004; Kern *et al*. 2009; Popa and Kern 2009) or drought stress (Levanic *et al*. 2009; Kane and Kolb 2010) can be ruled out as drivers of local TRD formation. TRDs are also largely absent in trees 3 and 4 sampled at the margin of the deposit at its height in 2009. Interestingly, TRDs exhibit the strongest concentration within the area previously covered by sediment whereas they occur with progressively reduced intensities (or are even completely absent) in the upper subaerial segments of the stem. Their distribution seems to confirm our assumption that the presence of TRDs is linked to the presence of toxic, sulphur-rich sediment around the stem base.

Conclusion

The Dumitrelul spoil heap was and still represents a source area of sulphur-rich material mobilization and continues to represent a threat for the Dumitrelul reservoir and the populations living downstream of the area. The vegetation affected by toxic sediments will continue to be affected by sediment transfers in the future and will eventually die. This study not only calls for more research on TRD formation in intoxicated trees, but mainly also for a rehabilitation of the area and in particular the spoil heap located upstream of the reservoir.

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