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Integrated biomonitoring of airborne pollutants over space and time using tree rings, bark, leaves and epiphytic lichens

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ABSTRACT

The integrated use of tree rings and outer tissues, and lichens, was tested for monitoring how pollutant concentrations vary in space and over time nearby an incinerator in industrial area in Central Italy. Trace elements in thalli of lichen *Xanthoria parietina* and in leaves, bark, wood of *Quercus pubescens*, as well as carbon, oxygen and nitrogen isotope ratios in tree rings were analyzed. Some trace elements in the leaves differed significantly between the plots, though this was not the case in lichens and bark. The values of $\delta^{13}C$ and $\delta^{18}O$ showed the same trend in all plots, while the values of $\delta^{15}N$ were higher in the distal plot. The results indicated that trace elements were intercepted and collected by tree bark and leaves, as well as lichens, at low concentrations, and that they hardly entered into tree xylem tissues during the growing season to be stored into the woody tissues. Indeed, the study did not highlight marked changes over time and space, in accumulation of airborne pollutants in the selected biomonitors, most probably due to the low levels of industrial development. Nevertheless, the analysis of tree ring cores in combination with bark and leaves, and lichens might potentially contribute to depict historic impacts of airborne pollutants at pronounced concentrations.

1. Introduction

Humans and environments are continuously exposed to pollutants via natural and anthropogenic processes. Although the evidence base is as yet weak, there is accumulating evidence that exposure to pollutants is associated with adverse effects on human health, affecting particularly the cardiovascular system (Heal et al., 2012). Plants also exhibit an evident symptomatology when exposed to phytotoxic concentrations of pollutants with altered growth, physiology and reproductive patterns, or productivity and distribution (e.g., Lin and Xing, 2007). However, urban vegetation may efficiently remove airborne pollutants simply as a function of deposition velocity, pollution concentration, and tree structure (biomass) (e.g., Chen et al., 2015). Urban and peri-urban trees are considered, therefore, providers of ecosystem services and monitoring tools; these biomonitors give quantitative or qualitative indications of pollution (Wolterbeek, 2002). For example, leaves have been studied to assess the effects of atmospheric pollution, and sensitive species can be used as biomonitors of trace elements (e.g., Tomăsevíc et al., 2011). The concentration of essential and non-essential elements in leaves provides information on the incidence of each element in the environment (Ugolini et al., 2013), since temporal trends in element accumulation of leaves can be consistent (Aničić et al., 2011). Bark also accumulates atmospheric particulate matter in its outermost surface (e.g., Catinon et al., 2011). However, the mechanisms and physical-chemical characteristics of the deposition of elements of atmospheric origin and their accumulation in bark remain poorly understood (Suzuki, 2006).

Tree rings have been used to monitor stress-induced changes in trees (Savard, 2010), providing signals of past pollution in areas where the monitoring of emissions has been short term or not existent (Ferretti et al., 2002). Tree rings provide information about chemical changes in the concentration of pollutants over time, as well as about how variations in atmospheric deposition affect some key physiological processes and how trees adapt under changing ecological conditions (Novak et al., 2007; Leonelli et al., 2012).

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Long-term tree-ring series represent useful archives, recording past environmental conditions and physiological responses, which are not always directly measurable in urban areas (Battipaglia et al., 2010). The analysis of tree rings for monitoring trace metal pollution is based on the element concentrations in the tree that, therefore, represents the temporal evolution of element availability in the environment in which the tree grows (Baes and McLaughlin, 1984; Padilla and Anderson, 2002; Doucet et al., 2012). Trees may accumulate anthropogenic pollutants either from the atmosphere, by deposition onto foliage and/or bark, and/or following deposition on the soil and subsequent uptake by roots. Pollutant-induced datable signals in tree rings can be useful to reconstruct environmental pollution over time.

Lichens have been widely used as bioaccumulators of trace elements, since their metabolism is very dependent on pollutant atmospheric exchanges and they are metal tolerant (Bargagli and Nimis, 2002). Lichens are able to accumulate trace elements in very high levels, and the concentrations of the trace elements inside the thalli of several lichen species seem to be directly correlated with the environmental levels of these elements (Paoli et al., 2013).

While several studies have focused on both lichens and mosses as bioaccumulators, which take up and retain metals differently (e.g., Szczepaniak and Biziuk, 2003; Giordano et al., 2013), few have used a multi-source approach. Some have compared lichens and bark (e.g., Santitoro et al., 2008) and lichens and leaves (e.g., Aprile et al., 2010), but none have considered combining lichen thalli and tree rings.

In this study, a novel approach to integrated biomonitoring has been developed that uses the chemical traits and ecophysiological processes in two different biomonitors, lichens and trees, in temporal and spatial combination. Bioaccumulation data on trace elements in leaves, bark, lichens and tree rings was used to evaluate the potential of this coordinated approach for monitoring the spatial and temporal impact of anthropogenic activities. Moreover, stable isotopes ratio of C, N and O in tree rings have been used as long-term and sensitive indicators of environmental factors, including pollutants, that might influence the plant functionality (e.g., Guerrieri et al., 2011). Changes in assimilation rate and stomatal conductance are detectable through the variation in stable isotope ratios of C, δ^{13}C, and O, δ^{18}O, respectively, from tree rings (Scheidegger et al., 2000; Saurer and Siegwolf, 2007). Whereas, NOx emissions and N deposition related to anthropogenic activities can affect N cycle, changing δ^{15}N signature in tree rings (Saurer et al., 2004).

The potential of the wood matrix to determine any possible dioxin contamination was also explored. Dioxins are known to be formed during the combustion of industrial and domestic waste, and to escape from incinerators into the environment via exhaust gases (Shibamoto et al., 2007). Tree rings may show environmental signals over time in reaction to different industrial activities, e.g., dioxin accumulation.

To test the validity of this monitoring protocol, hypothesizing that biomonitors would yield similar information, we gathered data on a line transect across an area affected by the emissions of a nearby incinerator plant. The aims of this study were: i) to assess whether pollutants are accumulated differently in lichens, leaves, bark and tree rings according to their specific sensitivities; ii) to determine whether the increasing distance from the incinerator plant affects pollutant accumulation profiles in biomonitors; iii) and to test whether temporal patterns recorded in biomonitors can be used to retrospectively reconstruct pollution disturbance (dendrochemistry for long-term interval, lichens, bark, and leaves as annual indicators).

2. Materials and methods

2.1. Site description

The study area is located in the Venafro Plain (Molise, Central Italy; 41°29′ N, 14°03′ E, 222 m a.s.l.). The climate is temperate sub-continental, with mean annual temperatures in the range of 10–14 °C and precipitation around 700 mm, N–NE and SW winds prevail throughout the year.

The local industry consists of about 30 small companies, which manufacture and process metals, chemicals, plastics, electronics and agri-food products, among other things, there is also a municipal solid waste incinerator, combined with an electric power plant for energy generation from waste burning. Data on the deposition of some airborne pollutants during the study period were available from ARPA Molise (2012): NO_2 deposition varied between 8 and 26 $\mu g\, m^{-3}$, SO_2 0.1–1.4 $\mu g\, m^{-3}$, PM_{10} 20–43 $mg\, m^{-3}$, NH_3 1–10 $\mu g\, m^{-3}$, As, Cd and Ni < 0.5 $ng\, m^{-3}$ and Hg < 0.001 $ng\, m^{-3}$.

The incinerator of Venafro, located NE of the town, has been in operation since 1997 until 2005. It mainly incinerated nut residues, mostly walnut, as well as residues from other fruit including those from olive oil production. The incinerator has diversified its activity in the last 15 years, and currently produces energy from the combustion of Refuse Derived Fuel (RDF).

The airborne emissions from the incinerator are continuously monitored instrumentally (http://www.energonut.it/emissioni_online.php). Punctual measurements of emissions since 2010 have consistently found values lower than the limits for pollutants specified in the national law (Italian Legislative Decree 133/2005).

2.2. Sampling

We aimed at detecting pollutant signals in different biomonitors, taking into account the different length of exposure in the environment, i.e., seasonal in leaves (sampled at the beginning and end of the vegetative season in 2012), a few years in lichen thalli (every 4 years), soil and bark (2012), and long term (several years) in wood. To compare the biomonitors, 2012 was considered a representative year for the completeness of available information, including on the deposition of airborne pollutants (ARPA). Three sampling plots were located at 380 m from each other along a transect from the incinerator to NE (Fig. 1) according to: (i) the localization of the solid waste incinerator, (ii) the dispersion model of PM_{10} (see Buonanno et al., 2010), assuming vegetation as the recorder of air pollution, (iii) the wind frequency distribution (data from: http://energonut.it), (iv) the horography and urbanization of the neighborhood, and (v) the two hilly chains bordering the area from NE to SW. Elevation ranges from 200 m in the lower area up to 900 m in the surrounding hills.

Soil samples (n = 3) in the form of three sub-samples per plot were collected. These were taken from the upper 5 cm of soil, the soil layer most sensitive to changes in atmospheric deposition (Augusto et al., 2010). The samples were sieved through a 2 mm screen, transferred to glass bottles to prevent adsorption by plastic, protected from sunlight and stored at 4 °C. The biological samples (leaves, bark, wood and lichen thalli) were collected from the same replicate trees (n = 3, each constituted by 6 sub-samples; n = 6 for dendrochemistry) in each plot. Healthy leaves of *Quercus pubescens*, the dominant trees in the local forest ecosystem, were collected in May at the beginning and in November at the end of the growing season 2012. Leaves were not washed in order to determine the total deposition, such as small particles incorporated into epidermal leaf structures and large agglomerates trapped onto the surface wax (e.g., Schreck et al., 2012).

Bark samples of *Q. pubescens*, in the form of a thin layer (<1 mm), were collected in three sub-samples at the end of June, after two