

## Black SOM dynamics during reforestation of Japanese grassland

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The dynamics of the polyaromatic structures of black humic acids (HAs), which are presumably derived from charred materials, are of significant interest for the global carbon cycle. However, the details of those dynamics are not yet well understood. We investigated differences in the degree of darkness (A<sub>600</sub>/C values), isotopic ratios ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ , and  $\Delta^{14}\text{C}$  values), and <sup>13</sup>C NMR spectra of size-separated black HAs extracted from Japanese volcanic ash soils in order to estimate the variations in the polyaromatic structures of black HAs during ca. 100 years of natural reforestation of Japanese pampas grassland. For several hundred years, all the study sites were managed similarly as grassland by burning. Subsequently, their management differed: at site G (*Miscanthus sinensis*: C<sub>4</sub> plant), maintenance as of the time of this study was still performed by mowing, while at sites P (*Pinus densiflora*: C<sub>3</sub> plant) and Q (*Quercus crispula*: C<sub>3</sub> plant), maintenance was discontinued ca. 30 and 100 years ago, respectively. Thus, the sites range from grassland (site G) to coniferous forest (site P) to broad-leaved forest (site Q). For all HA size fractions at all sites, we found that  $\delta^{13}\text{C}$  values correlate positively with  $\delta^{15}\text{N}$  values, although the gradients are much lower for fractions of small to medium molecular size than for fractions of medium to large molecular size (denoted as lower-size and higher-size fractions, respectively). Overall, for the lower-size fractions, the contribution ratio of C<sub>4</sub>-plant-derived carbon shows a significant positive correlation with A<sub>600</sub>/C values and a negative correlation with  $\Delta^{14}\text{C}$  values, and their aromatic characteristics are greater than those of higher-size fractions within the same black HA. Furthermore, the relative proportion of lower-size fractions decreases with reforestation, especially from site P to Q. The  $\delta^{13}\text{C}$  values for all size fractions are similar for sites G and P, but are relatively low for site Q. The aryl C contents of the lower-size fractions are lower and the O-alkyl C contents and the aliphaticity (alkyl C:O-alkyl ratio) are clearly higher for sites P and Q than for site G. These results strongly suggest that stimulation of HA biodegradation might be achievable by continuous input of new plant litter during reforestation, even for lower-size HA polyaromatic structures, despite the fact that lower-size HAs biodegrade more slowly than higher-size HAs.

Keywords: land use, reforestation, soil organic matter, <sup>14</sup>C