

Figure S1. Carbon (C) biomass per unit ground area simulated by LPJ-GUESS. Spatial distribution in the PNV scenario (a). Panel b-c show changes in the C mass from the deforestation (DCGL–PNV) and reforestation (DCGL–DCL).



Figure S2. Changes in the tree and grass cover fractions (area of vegetation per unit ground area) from reforestation (DCGL–DCL). The right panels show the tree PFTs while the left panels show grass PFTs.



Figure S3. Total column bSOA from PNV (a) and changes in bSOA from deforestation (DCL – DCGL) (b). Panels (c), (d), and (e) show the vertical profiles of bSOA, represented by box-whisker plots for different pressure bins. The white line marks the median, the box corresponds to the lower and upper quarterlies, and the whiskers represent the 5th–95th percentile. The latitude ranges are defined as follows: High latitudes (90–55°S and 55–90°N), mid-latitudes (55–30°S and 30–55°N), and low latitudes (30°S–30°N). A log scale is used for the x-axis of (c), (d), (e).



Figure S4. Spatial maps of the total column aerosol optical thickness (AOT) at 550 nm for OC (a), and total aerosol (e). Vertical profiles for the aerosol extinction at 550 nm for DCL, DCGL, along with the relative differences ((DCL–DCGL)/DCGL) in different latitude bands. Organic aerosol in (b), (c), (d), and total aerosol in (f), (g), (h). The latitude ranges are defined as follows: High latitudes (90–55°S and $55-90^{\circ}$ N), mid-latitudes ($55-30^{\circ}$ S and $30-55^{\circ}$ N), and low latitudes (30° S– 30° N). The grey area represents 1 standard deviation of the spatio–temporal mean (grey line). Please note the different scales for the relative differences.



Figure S5. Changes in cloud condensation nuclei (CCN) at 0.2% supersaturation (top panels) and cloud droplet number concentration (CDNC) (bottom panels). The panels on the left-hand side, (a) and (e), show the spatial difference in the total column burden (number of particles per square meter), for CCN and CDNC, respectively, emerging from DCGL compared to natural vegetation (DCL – DCGL). The panels on the right-hand side show the total-column vertical profiles from DCL and DCGL simulations and their relative difference are shown.



Figure S6. Relative changes in atmospheric states resulting from present-day land use cover compared to natural vegetation cover. Maps show variations in; (a) bSOA column mass, (b) total aerosol optical thickness (AOT), (c) cloud condensation nuclei (CCN), (d) cloud droplet number concentration (CDNC), (d) aerosol radiative effect, and (e) cloud radiative effect.



Figure S7. Relative changes in atmospheric states resulting from restoring present-day grazing land. Maps show variations in; (a) bSOA column mass, (b) total aerosol optical thickness (AOT), (c) cloud condensation nuclei (CCN), (d) cloud droplet number concentration (CDNC), (d) aerosol radiative effect, and (e) cloud radiative effect.