Supporting Information
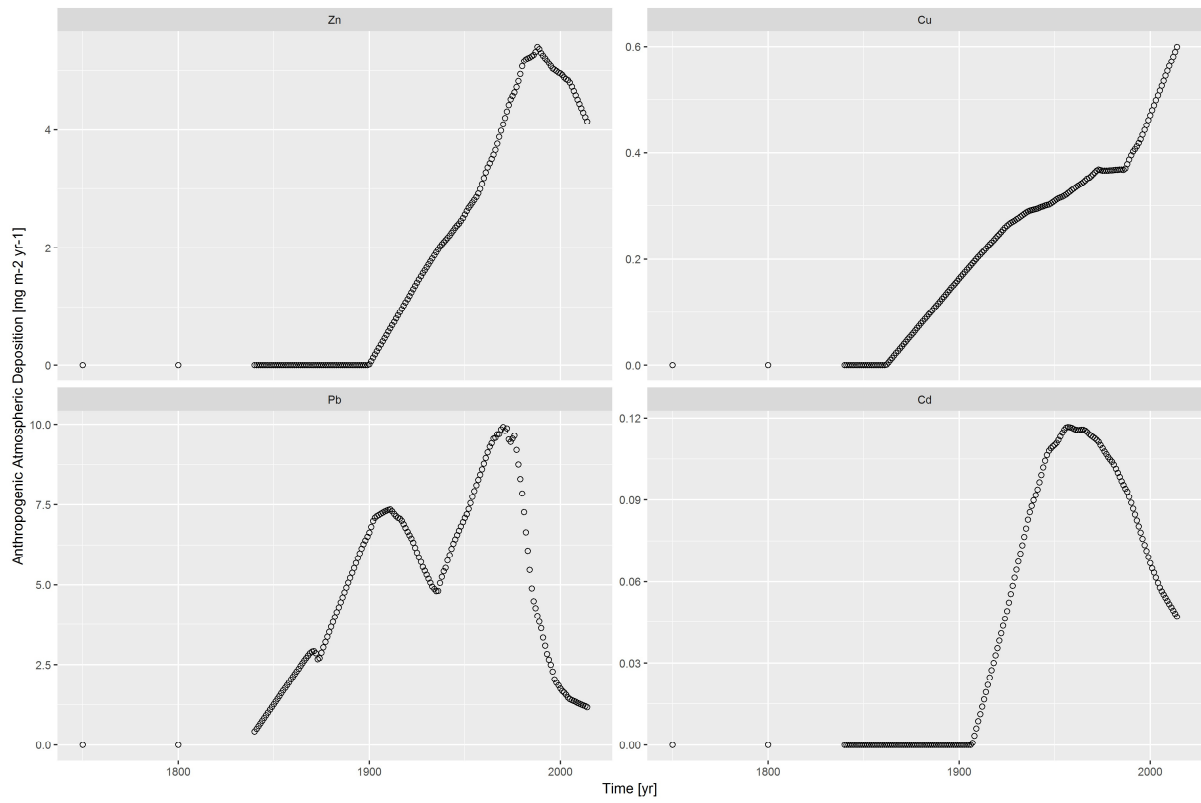
Figure S1: Estimated input fluxes of Zn, Cu, Pb, Cd via anthropogenic atmospheric deposition.
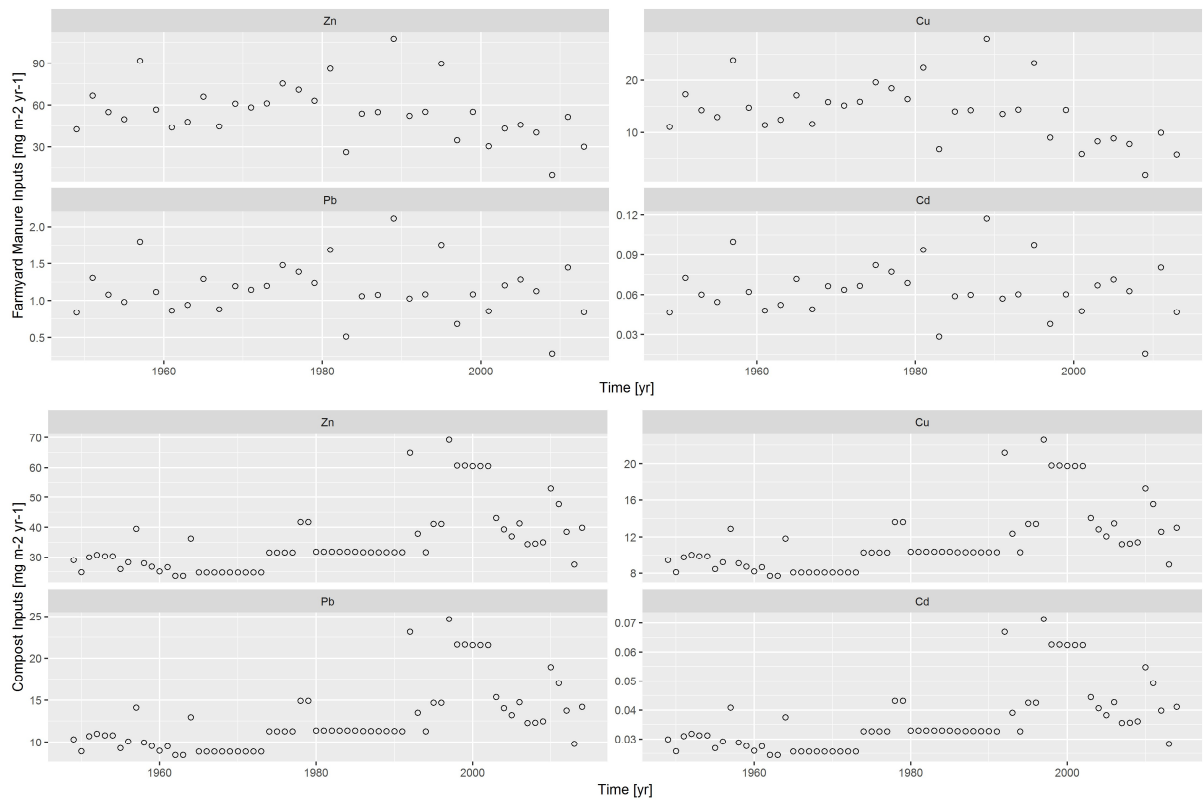
Figure S2: Estimated input fluxes of Zn, Cu, Pb, Cd via application of farmyard manure and compost amendments.
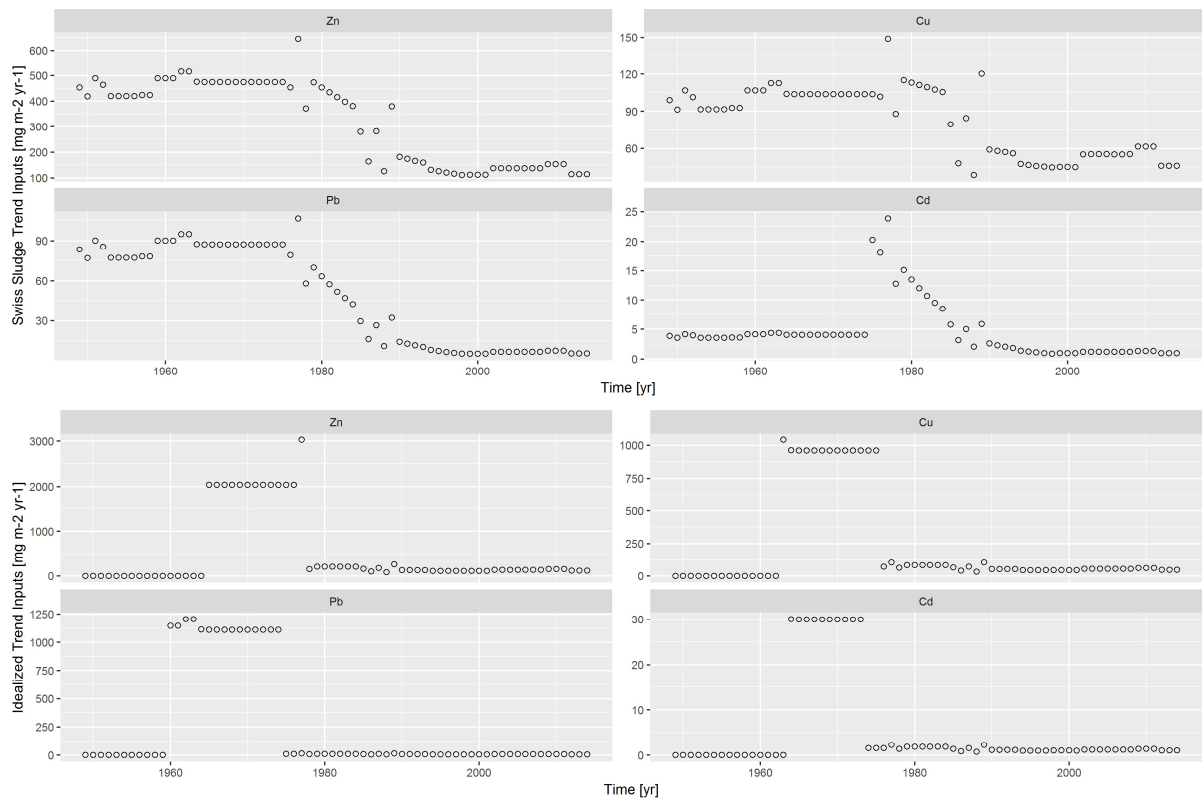
Figure S3: Estimated input fluxes of Zn, Cu, Pb, Cd via application of sewage sludge amendment according to the *'Swiss Sludge Trend'* approach (top) and *'Idealized Trend'* approach (bottom).
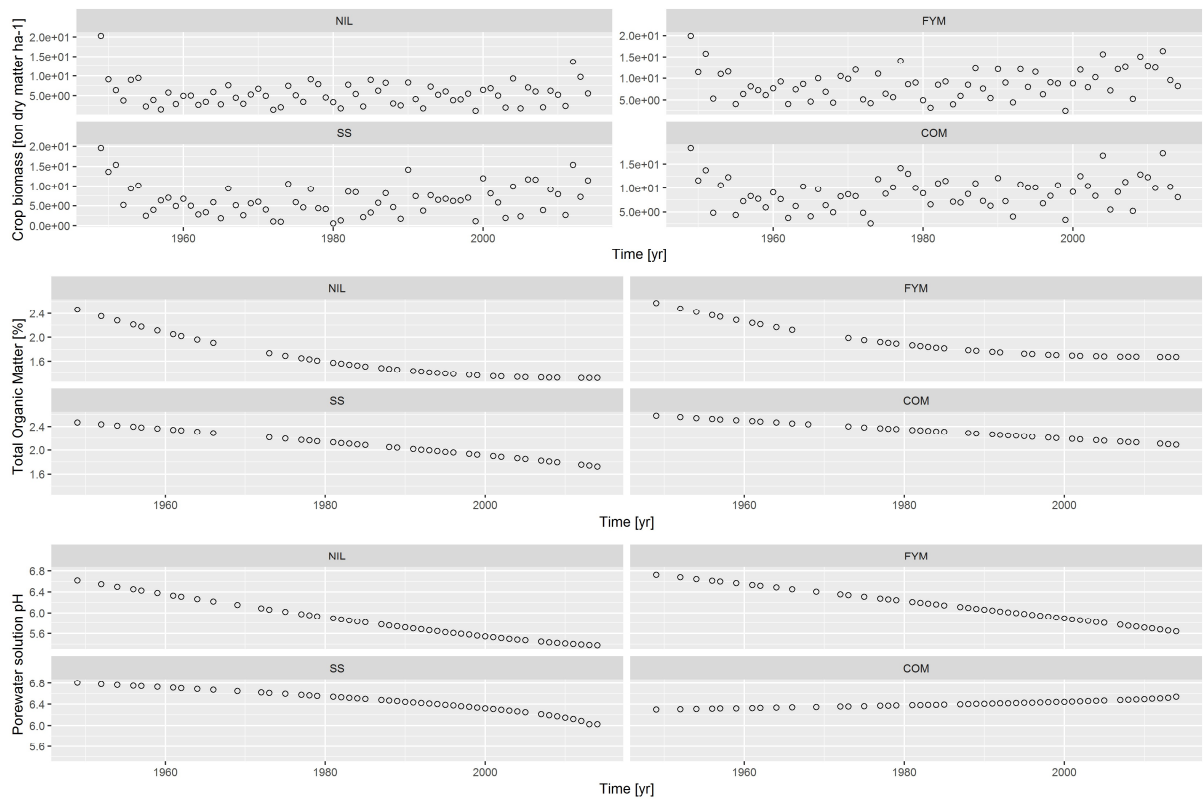
Figure S4: Estimated crop biomass, total organic matter and soil solution pH in the treatments NIL, FYM, SS, COM.

The most noticeable peaks in the FTIR spectra were attributed as follows: $3624\,cm^{-1}$ (Al–OH) vibrations, probably caused by fine material like clay (Cl); $2920\,cm^{-1}$ and $2850\,cm^{-1}$ (aliphatic groups asymmetric and symmetric –CH stretching, respectively), (OM_1); $1850\,cm^{-1}$, $810\,cm^{-1}$ $696\,cm^{-1}$ quartz (Q_1, Q_2, Q_3,); $1630\,cm^{-1}$ (aromatic –C=O stretch and/or asymmetric of –COOH); $1650\,cm^{-1}$ and $1550\,cm^{-1}$ [amide I (–C=O of amide group) and amide II (N–H bending), respectively], (OM_2); $1460\,cm^{-1}$ (–CH deformation of CH_3), (OM_3); and $1040\,cm^{-1}$ (–C–O stretching of polysaccharide or polysaccharide-like) (OM_4).
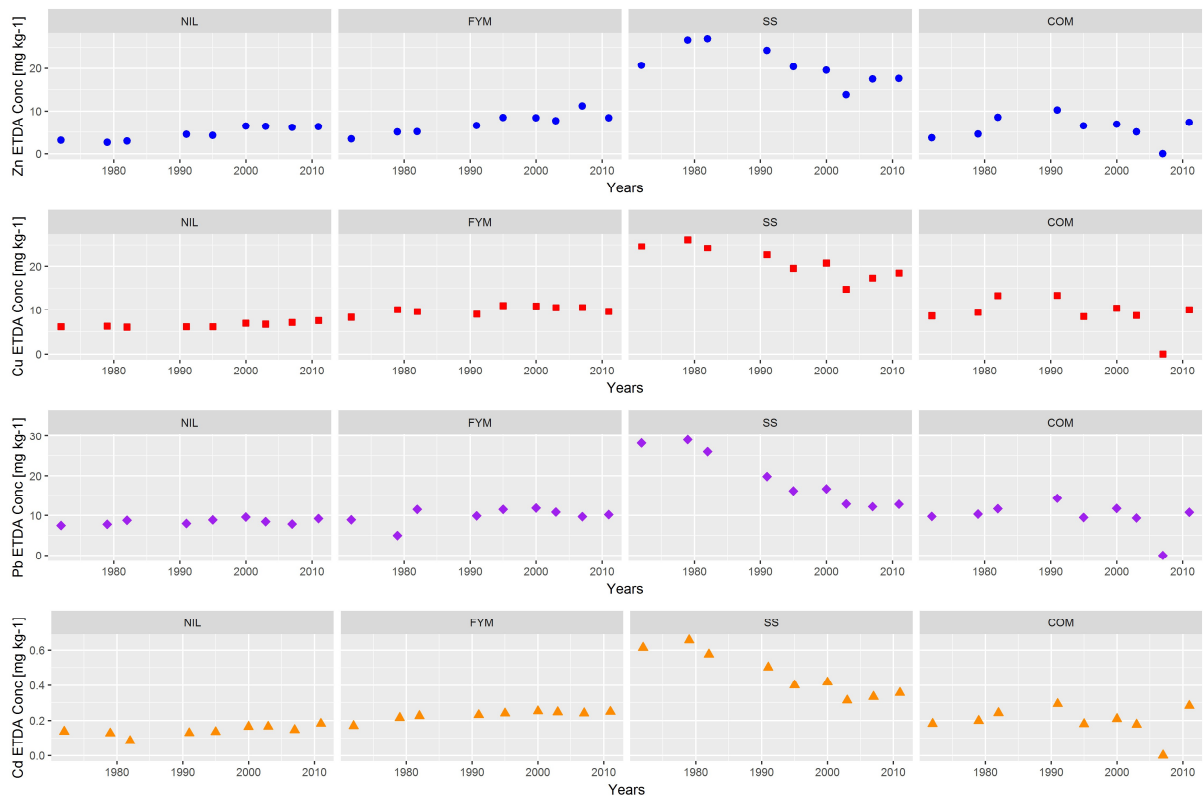
Figure S5: EDTA-extractable concentration time trends of Zn, Cu, Pb and Cd in ZOFE topsoils (20cm) for the treatments NIL, FYM, SS and COM.
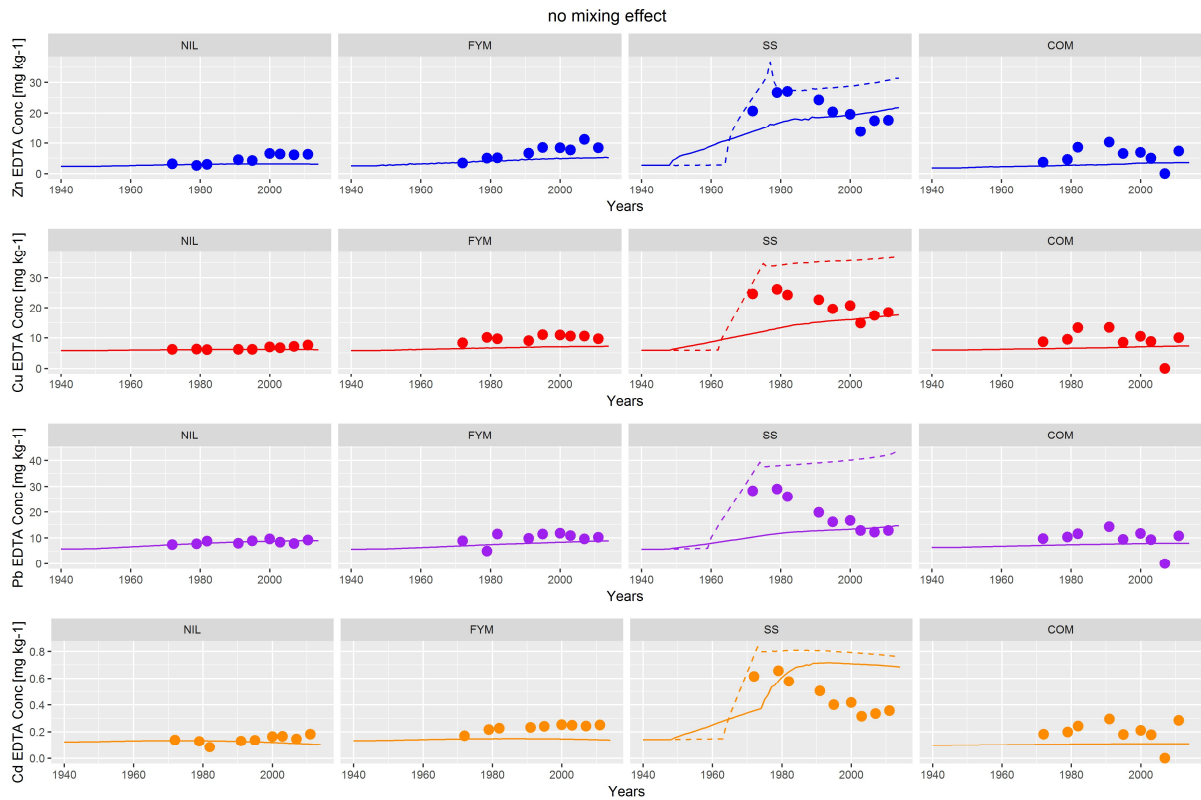
Figure S6: Measured EDTA-extractable concentrations (•), simulated labile pool concentrations with the "Swiss Sludge Trend" approach (—), simulated labile pool concentrations with the "ZOFE Sludge Trend" approach (--) of Zn, Cu, Pb, Cd in ZOFE topsoils (0-20cm) for the treatments NIL, FYM, SS and COM. All the simulations are carried out without any lateral mixing effect. The two approaches differ in the way the sewage sludge metal content is estimated over time.
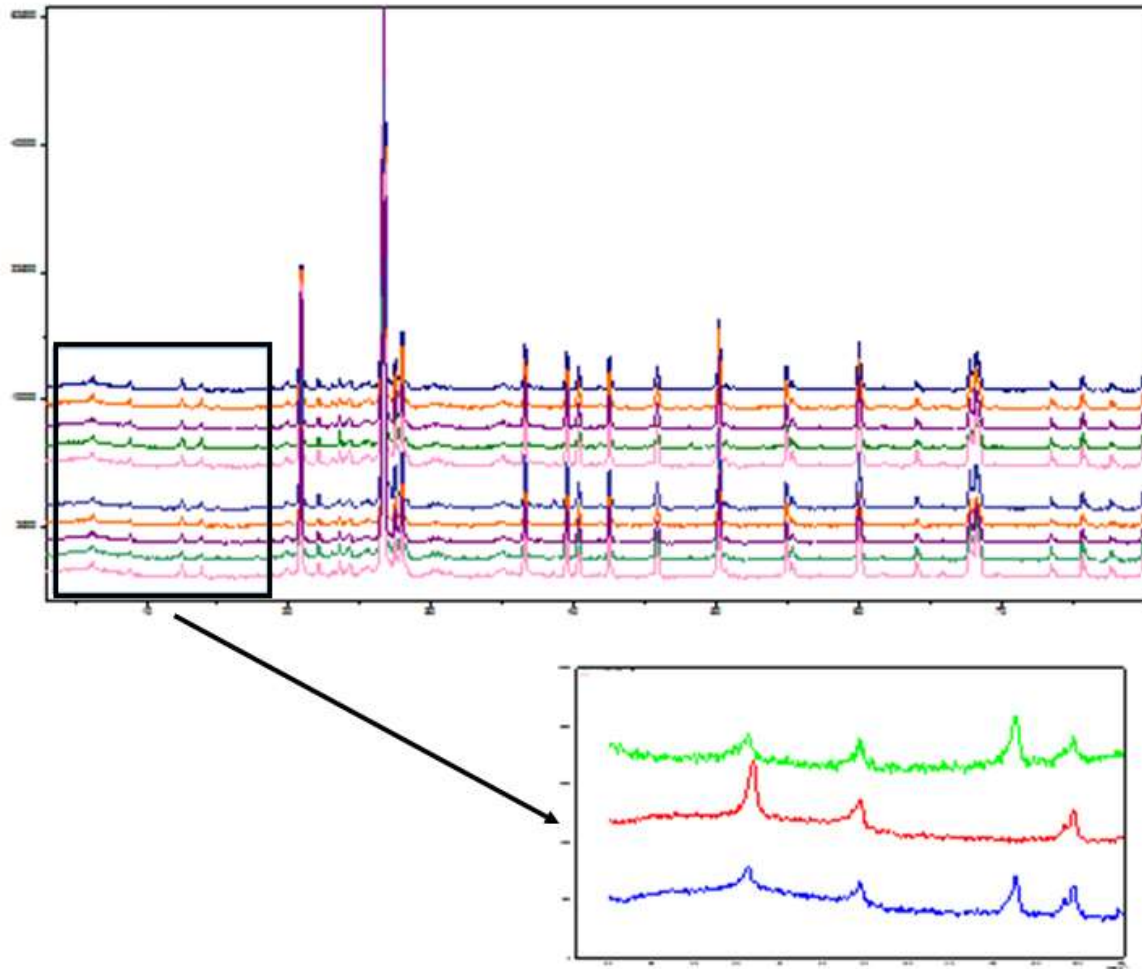
Figure S7: XRD diffractograms of NIL, FYM and COM soil samples from 1972 and 2011 and SS samples from 1972, 2011 and 2013 in ZOFE topsoil (0-20cm).