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Characterization of organic acids, carbon and nitrogen in rainwater and aerosols in Brazil: from natural biogenic emissions through anthropogenic impacts.

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This paper describes the relationship between biogenic emissions from forests of Amazon Basin and Southeast Brazil and the concentration of organic acids in rainwater and aerosols. We also addressed how the anthropogenic activities are changing the concentration and deposition patterns of organic acids, carbon and nitrogen in Brazil. The atmospheric deposition of chemical species within the Earth's ecosystems not only provides a natural sink but also acts as a source of nutrients and plays an essential role in controlling the biogeochemical cycles of carbon and nitrogen. In tropical regions convective clouds produce abundant rainfalls which are the main sink of water-soluble gases and aerosols emitted into the atmosphere through wet deposition processes. In the tropics, the intense land-use changes followed by rapid urbanization, associated with a large industrial expansion are possibly altering the atmospheric chemistry. Indeed, the acidification of precipitation and subsequent high nitrogen deposition rates has been taken place in disturbed regions in Brazil.

In order to investigate the relationship between land cover and atmospheric chemistry we collected rainwater and aerosol (fine and coarse mode) samples in different sites in Southeastern Brazil and in Amazon Basin. Pristine sites are located in Central Amazon rain forest (Balbina) and in Mata Atlântica (native forest) in Southeastern Brazil. Disturbed sites are characterized by deforestation and biomass burning (Rondônia, Amazon Basin), industrialization and urbanization processes and biomass burning

(Southeastern Brazil). Land cover and land-use changes are probably responsible for the spatial variability found in precipitation and aerosol chemistry. As a consequence of the anthropogenic activities significant rainfall acidity was detected (VWM pH = 4.5). The origin of the free acidity in rainwater is different in each site and come out to be linked to the land cover. Organic acids appear to control the acidity in remote areas while in other sites inorganic acidity has been detected. In addition, land-use changes, mostly the conversion of primary forest to pasture or croplands by biomass burning, are altering the concentration and the pattern of organic acids in rainwater.

According to the Factor and Cluster Analyses, the composition of rainwater and aerosols in the disturbed sites appears to be controlled mostly by two sources: biomass burning and industrial emissions, and in the remote areas by biogenic emissions and marine influence. Additionally, nitrogen deposition was significantly higher in the disturbed sites and DOC (Dissolved organic carbon) deposition was significantly lower in these sites. Indeed, N wet deposition increases from an annual rate of 3.0 $kg.N.ha^{-1}.yr^{-1}$in pristine areas to an annual rate of 5.6 $kg.N.ha^{-1}.yr^{-1}$ in disturbed regions. Our results pointed out that this increasing is mostly linked to biomass burning emissions which also drive a shift in the composition of nitrogen deposition from nitrate to ammonium dominated nitrogen deposition budget. Moreover, the pattern of organic acids such as the rate of formate to acetate also has been changed due to biomass burning. These large perturbations to the tropical atmospheric nitrogen cycle and organic acids have important deleterious consequences for ecosystem function. If the anthropogenic activities continues to accelerate at the present rate and if these changes in the atmosphere composition detected in Brazil are also taking place in other tropical areas of the world, where most of the biomass burning occur, in a few decades significant regional biogeochemical cycles changes can be expected with alterations in the regional climate and possible global impacts.