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Cloud chemistry at the Puy de Dôme: variability and relationships with environmental factors

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Abstract. The chemical composition of cloud water was investigated during the winter-spring months of 2001 and 2002 at the Puy de Dôme station (1465 m above sea level, 45°46′22′′ N, 2°57′43′′ E) in an effort to characterize clouds in the continental free troposphere. Cloud droplets were sampled with single-stage cloud collectors (cut-off diameter approximately 7 µm) and analyzed for inorganic and organic ions, as well as total dissolved organic carbon. Results show a very large variability in chemical composition and total solute concentration of cloud droplets, ranging from a few mg l^{-1} to more than 150 mg l^{-1}. Samplings can be classified in three different categories with respect to their total ionic content and relative chemical composition: background continental (BG, total solute content lower than 18 mg l^{-1}), anthropogenic continental (ANT, total solute content from 18 to 50 mg l^{-1}), and special events (SpE, total solute content higher than 50 mg l^{-1}). The relative chemical composition shows an increase in anthropogenic-derived species (NO_3^-, SO_4^{2-} and NH_4^+) from BG to SpE, and a decrease in dissolved organic compounds (ionic and non-ionic) that are associated with the anthropogenic character of air masses.
We observed a high contribution of solute in cloud water derived from the dissolution of gas phase species in all cloud events. This was evident from large solute fractions of nitrate, ammonium and mono-carboxylic acids in cloud water, relative to their abundance in the aerosol phase. The comparison between droplet and aerosol composition clearly shows the limited ability of organic aerosols to act as cloud condensation nuclei. The strong contribution of gas-phase species limits the establishment of direct relationships between cloud water solute concentration and LWC that are expected from nucleation scavenging.

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