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Aerosol pH 25-years trend predicted from fog composition in Po Valley, Italy

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pH is a fundamental aerosol property that affects ambient particle composition, concentration and toxicity, linking pH to all aerosol environmental impacts. Direct measurement of aerosol pH is highly challenging, and so indirect proxies are often used to represent particle acidity. Aerosol thermodynamic models, such as ISORROPIA-II, are able to calculate particle pH – based on concentrations of various aerosol species, temperature (T), and relative humidity (RH) – and offer a rigorous approach to obtain aerosol pH already tested in the past with ambient aerosol data. However not many long aerosol measurements datasets exist to understand the trend of particle acidity along the past decades in Europe as well as around the world. Long-term monitoring programs for cloud/fog composition and acidity are also lacking in the global scientific community, but there are a few locations around the world where such measurements have been made routinely or periodically over periods of a decade or more. One of these locations is the rural station of San Pietro Capofiume in the Po Valley (Italy), where a consistent long dataset of fog-water ionic composition exists spanning the last 25 years (1993-2018).

In this study, assuming that fog acts as an efficient natural scavenger of aerosol particles, we use the inorganic composition of fog-water collected at SPC as a proxy for the chemical composition of atmospheric aerosol in pre-fog conditions. So, we apply ISORROPIA-II to calculate the pH associated with particles having the same chemical composition of fog-water. In this way we extend the analysis to the long-time record of fog-water measurements obtaining the aerosol pH trend of the last 25 years. A comparison with existing aerosol samples and parallel ammonia gas measurements allow us to validate the approach.

Our thermodynamic analysis suggests a decreasing trend of aerosol pH in Po Valley. Over the twenty-five-year period the aerosol pH decreased approximately 1.1-1.6 pH units, progressing also with an increasing rate of reduction, which corresponds to 0.18 pH units between the first and the

second decades (1993-2002 and 2003-2012 respectively) and 0.44 between the decade 2003-2012 and the last 6 years (2013-2018).

A multiple linear regression analysis applied on the simulated aerosol pH reveals that the aerosol pH reduction trend is driven by the contemporary decrease of the main pollutants atmospheric concentration (possibly due to the European environmental policies) and by the changing meteorogical parameters (T and RH), possibly linked with climate change.

Our analysis suggests for the first time the possibility of calculating pre-fog aerosol pH using fog compositional data in a thermodynamically consistent way, which can be useful to evaluate long-term trend of particles acidity also in other region of the world for which data are available (e.g., Californian Central Valley).

Projecting the trend in the future it is possible to speculate a potential change in deposition of nitrate/nitric acid from aerosol-dominant (slow) to gas-dominant (fast) with very important consequences in air quality.

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