Atmospheric aerosol nucleation in the Po Valley during the PEGASOS-SUPERSITO experiment

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Aerosol nucleation, or new particle formation (NPF), is a very common phenomenon in the atmosphere, exerting an important feedback on the climate system by sustaining cloud condensation nuclei (CCN) concentrations in areas relatively far from pollution sources. In forest areas, NPF was put in relation to biogenic VOC photochemistry, while the mechanisms triggering nucleation in more anthropized environments are more elusive. North Italy is exemplificative of such an environment, with forested mountain ridges and urban and rural areas extending over the low lands (the Po Valley). During the PEGASOS-SUPERSITO field campaign in June-July 2012, the frequency of NPF in the rural Po Valley was very high (87% of the days) compared to the Apennine mountain ridge (35%). At the low-elevation rural site, nucleation started in the first two hours following sunrise, before the mixing layer development, in an atmospheric layer characterized by reduced ozone concentrations, high NOx, relatively high anthropogenic VOC levels (200 ppt of toluene), ppt-level of isoprene, and moderate SO_2 concentrations (1 ppb). Clearly, the pool of possible precursors for nucleating particles in the Po Valley was dominated by anthropogenic species. Finally, the combination of the observations carried out at the ground stations with simultaneous measurements performed with mobile platforms provided evidence of the fact that NPF in the Po Valley was not driven by emissions from point pollution sources (such as power plants) but occurred in the background air over vast sectors of the valley. Continuing developing experimental and modelling tools to understand the subtle mechanisms underlying new particle formation will greatly improve our ability to assess the impacts of such pervasive aerosol source at the global and regional scales.

KINETIC14 : A PHREEQC compatible mineral kinetic database

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Developped within the CarbFix project [1], KINETIC14 is an add on to PHREEQC [2], allowing calculation of the temporal evolution of mineral-fluid reactions during natural geochemical and industrial processes, and laboratory experiments.

Building upon previous efforts [3,4] this KINETIC14 has been built from a detailed synthesis of dissolution and precipitation rate data available in the literature. When coupled with PHREEQC, KINETIC14 provides the dissolution rates for 98 minerals as a function of temperature, fluid phase composition, and the chemical affinity of the fluid phase with respect to the mineral phase of interest. Mineral-fluid interfacial surface areas required for modelling the evolution of the fluid phase with these rates can be calculated using a variety of geometric models or from user input.

The rate equations chosen to fit available data within KINETIC14 are based on transition state theory and dissolution mechanisms that take explicit account of reactions occuring on mineral surfaces, including metal for proton exchange reactions, multi oxide minerals and proton adsorption/desorption for simple oxides. Despite the substantial scatter that is pervasive in dissolution rate data reported in the litterature, the majority of fitted rates are within 0.8 log units of their experimentally measured counterparts.

Mineral precipitation rates are generated directly from the corresponding dissolution rate equations; the validity of this approach will need to be assessed with future research.

Upon final validation, KINETIC14 will be made publicly available.

[1]Matter *et al*.. (2011) *Ener. Proc.***4** [2]Parkhurst and Appelo (1999) *USGS* [3] Marini (2007) *Dev. Geochem.* **11** [4] Palandri and Kharaka (2004) *U.S.G.S.,Open File Report*

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