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An overview of results on physico-chemical processes obtained from the MISTRALS/ChArMEx program

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This paper gives an overview and synthesis over results on chemical and physico-chemical processes obtained within the MISTRALS/ ChArMEx program. Indeed, one of the objectives of ChArMEx was to study and assess formation processes of secondary pollutants over the Mediterranean basin. Although these processes are fundamental, conditions over the basin are specific, with a large variety of sources mixing (anthropogenic and biogenic continental, shipping, marine, mineral dust...), long processing times, and intense actinic fluxes leading to intense photo-chemical activity. Results were obtained in particular from intensive measurement campaigns during summers 2013 and 2014 over the western Mediterranean basin, combining ground-based, balloon-borne, ship and aircraft measurements. We focus here on several targeted physico-chemical processes:

- Reactivity and functionalization of organic matter.
- Radical budget including heterogeneous reactions impacting on it.
- New particle formation.
- Formation and evolution of organic aerosol particles.
- Photochemical ozone production.

New particle formation by nucleation at large scale and at various altitudes was made evident from ambient measurements, and their possible association to the transition between polluted and clear air masses shown. In addition, the role of iodine compounds of marine biogenic origin in the formation of new particles was demonstrated from mesocosm experiments. A first high resolution multi-year monitoring of fine particle chemistry in the western Mediterranean at Cape Corsica showed the very oxidized state of organic aerosols in all seasons, but most pronounced in summer, and the large contribution of biogenic sources. Air quality models with updated organic aerosol schemes and improved chemistry are able to reproduce organic aerosol concentrations and its split into a fossil and modern (biogenic) fraction. A sensitivity analysis showed the possible benefit on air quality of reducing ship emissions in particular in the Gulf of Genova and Ligurian Sea. BVOCs control the main part of the calculated OH reactivity. Nevertheless, a significant missing reactivity is observed during some periods. New efficient mechanisms of SO_2 to H2SO4 conversion by Criegee intermediates recently made evident in laboratory experiments were found of probable minor importance in the field conditions of Cape Corsica in summer. For semi-volatile organic compounds, equilibrium is often shifted to the particulate phase with respect to partition theory, which suggests inhibition of evaporation of these compounds by a solidified particle surface, or likewise loss reactions of these compounds in the particulate phase. From drifting balloons over the basin, rates of O_3 photochemical formation could be estimated in the marine boundary layer and even in the free troposphere.