Spanish activities in the framework of the ChArMEx project since 2009: a summary

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Abstract — The ChArMEx (Chemistry-Aerosol Mediterranean Experiment) project is a French initiative aiming at developing and coordinating regional research actions for a scientific assessment of the present and future state of the atmospheric environment in the Mediterranean Basin, and of its impacts on the regional climate, air quality, and marine biogeochemistry. The target of ChArMEx is short-lived particulate and gaseous tropospheric trace species. In 2009 the project gained internationalization with the organization of the first international workshop held in Toulouse, France. Spain was the most represented country (after France) with 7 groups and 10 researchers. Up to date, the Spanish groups involved in ChArMEx have conducted research in several fields. Among them it is worth noting: a 3-year (2010 – 2012) in-situ study over a regional background environment in Mallorca plus 3 intensive measurement campaigns in that period; the installation in Mallorca and in the Sierra Nevada of two autonomous total deposition samplers performing weekly dust deposition on a network basis; and intensive in-situ and remote sensing measurements in Barcelona in the summer 2012 ChArMEx pre-campaign to give support to airborne observations. The paper gives a summary of all the activities led by Spanish researchers in the framework of ChArMEx. Preliminary results of the 3-year in-situ study in Mallorca and of the summer 2012 pre-campaign, as well as a brief description of the summer 2013 main campaign are presented.

Keywords — Aerosols, chemistry, atmosphere, climate, Mediterranean region, observations, remote sensing, modelling

1 INTRODUCTION

The international project ChArMEx (Chemistry-Aerosol Mediterranean Experiment) is a French initiative whose goal is a scientific assessment of the present and future state of the atmospheric environment in the Mediterranean Basin, and of its impacts on the regional climate, air quality, and marine biogeochemistry. The target of ChArMEx is short-lived particulate and gaseous tropospheric trace species. The major stake is an understanding of the future of the Mediterranean region in a context of strong regional anthropogenic and climatic pressures [1].

Through seven work packages, ChArMEx addresses: emissions and source apportionment; chemical ageing of air masses with a focus on the formation of secondary organic aerosols; transport processes and their effect on air quality; direct radiative forcing by aerosols and its consequences on the water budget and regional climate; atmospheric deposition of nutrients and contaminants; recent trends and variability in atmospheric composition; and the future evolution of atmospheric chemistry at the horizon of 2030 and 2050.

Since 2009 several Spanish centers are involved in the project, in particular in the workpackages emissions and source apportionment, direct radiative forcing and deposition. This paper concentrates on the Spanish activities led in those 3 workpackages. A brief description of the summer 2013 main field campaign is also presented.

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2 SPANISH ACTIVITIES WITHIN THE WORKPACKAGE 1: EMISSIONS AND SOURCE APPORTIONMENT

The most relevant contribution in WP1 is the implementation in an existing regional background monitoring site situated in Can Llompart in Mallorca Island (Fig. 1) of additional instrumentation to fulfil some of the following objectives: a complete chemical characterization of the atmospheric aerosols and the identification of their main sources; the assessment of the levels of gaseous pollutants and atmospheric aerosols in different particles sizes; and the quantification and chemical characterization of the deposition fluxes. The additional instrumentation was installed during a period of 3 years between 2010 and 2012. Three intensive measurement campaigns took place in spring and in summer 2011 and in summer 2012.

Fig. 1. Map of the Spanish sites involved in ChArMEx.

2.1 Preliminary results at Can Llompart

The instrumentation already existing at Can Llompart allowed measuring hourly levels of NO, NO_2, SO_2, O_3, PM_{10}, air temperature, wind direction and velocity, atmospheric pressure and precipitation. The additional instrumentation consisted of:

- an optical particle counter (OPC) to determine real time concentrations of PM_{10}, $PM_{2.5}$ and PM_1,
- two sequential high-volume (HV) sampler for PM_{10} and PM_1 collection on quartz micro-fibre filters,
- an automatic collector of dry and wet (D/W) deposition of atmospheric particles,
- an ultra-fine particle (5-1000 nm) counter for number concentration,
- a multi-angle absorption photometer (MAAP) for continuous monitoring of black carbon concentrations, and
- measurements of NH_3 concentration.

Further details about their period and mode operation are given in Table 1. In total over the period 2010-2012 more than 450 valid samples of PM_{10} and PM_1 have been collected. So far more than 400 of them have been analyzed for 60 inorganic compounds as well as organic and elemental carbon [2]. The PM_{10} daily concentration over the period 2010-2012 is shown in Fig. 2. The mean annual PM_{10} concentration over the whole period varies between 16 and 18 $\mu g{\cdot}m^{-3}$. All the daily PM_{10} peaks exceeding the limit value of 50 $\mu g{\cdot}m^{-3}$ are related to Saharan dust outbreaks. The average chemical composition over the whole period is formed dominantly by 23 % of organics, 17 % of mineral dust, 14 % of SO_4^{2-} (coal-fired power plant), 10 % of sea spray, and to a lesser extent nitrate and ammonium (5% each component). However, significant seasonal variations are observed for most of these components, especially for sulphate and mineral dust, clearly enhanced in summer, and nitrate, significantly higher in wintertime owing to the occurrence of NH_4NO_3 which is not stable under warm atmospheric conditions.

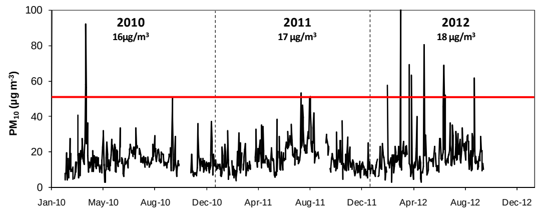

Fig. 2. PM_{10} concentration over the period 2010-2012 in Can Llompart.

2.2 Results of the 2011 field campaigns

The 2011 spring campaign (15 March – 28 April) was planned to capture the impact at ground level of long-range transported plumes in the western Mediterranean [3]. Several African dust episodes occurred in March and coincided with large amounts of anthropogenic, European pollution exported from the continent. During the campaign a severe PM pollution episode from Europe was captured, as well as typical summer-type African dust episodes [3]. Photochemical nucleation was frequently observed during the spring campaign, with strong peaks at midday coinciding with very low black carbon concentrations (Fig. 3). During such episodes, the number of ultra-fine particles (5-1000 nm) increased up to 120.000 cm^{-3} as hourly mean value 2 to 3 hours after the slight traffic peaks.

The 2011 summer campaign (21 June – 15 September) was intended to characterize in detail the alternate of regional recirculation episodes and African dust episodes affecting the western Mediterranean. Both types of episodes give rise to a clear increase of atmospheric pollution in the western Mediterranean, and favour the ageing processes of the atmospheric PM. Fig. 4 shows the PM_{1-10} fraction as well as the black carbon concentration. The accumulation of pollutants (regional recirculation) is indicated by upward red arrows in the BC plot, while the dust episodes, always coinciding with the end of the accumulation of pollutants, are indicated by black arrows in the PM_{1-10} plot.

The chemical analyses of PM_{10} and PM_1 samples obtained during both intensive campaigns, as well as the 2012 summer campaign, are currently ongoing.

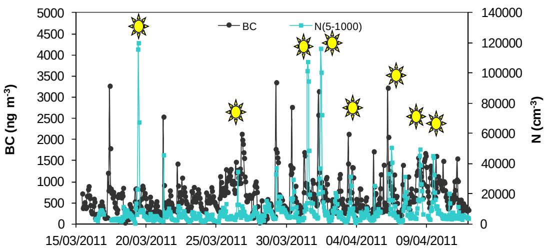

Fig. 3. Black carbon (BC) concentration and ultra-fine particle number (N) during the 2011 spring campaign in Can Llompart.

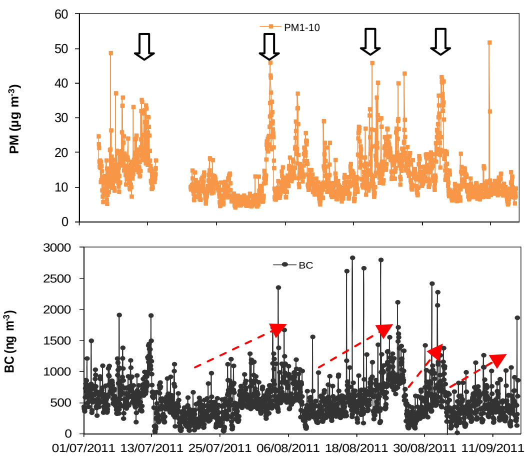

Fig. 4. Black carbon and PM_{1-10} during the 2011 summer campaign in Can Llompart.

3 SPANISH ACTIVITIES WITHIN THE WORKPACKAGE 4: RADIATIVE FORCING AND CLIMATE IMPACT

In the context of the 2012 ChArMEx pre-campaign, two field campaigns took place: TRAQA (Long distance transport and air quality in the Mediterranean Basin) and VESSAER (VErtical Structure and Sources of AERosols in the Mediterranean Region). The Spanish participation was mostly involved in the TRAQA campaign (25 June – 12 July). The main objectives of the campaign were [4]:

- Characterization of dynamic processes of exportation of contaminated air masses from source regions around the Mediterranean coast,
- Quantification of the exchanges between the boundary layer and the free troposphere above the Mediterranean Basin,
- Lagrangian track of the ageing and mixing of the contaminated plumes (aerosols and gases) in the low troposphere, and
- Analysis of the representativeness of the case study and the long-term series to evaluate the impact of the contaminated plumes on the air quality.

The meteorological conditions allowed for a total of 7 Intensive Observation Periods (IOP) with ~60 hours of flight of the instrumented ATR-42 research aircraft.

Intensive lidar measurements were performed in Barcelona to give support to the Saharan dust outbreak IOP (29 June), the pollution recirculation IOP (3-4 July) and the contamination structure along the Mediterranean coast IOP (10-11 July). Can Llompart measurements were intensified during the period 6 June – 15 August 2012.

In order to give a flavour of the ongoing analyses, Fig. 5 gives an illustration of the pollution recirculation IOP that took place in the area of Barcelona on 3 and 4 July. The MACC (Monitoring Atmospheric Composition and Climate) model first predicted an accumulation of pollutants in the area of Barcelona on 3 July that would travel northward on 4 July. The flights were set up accordingly to the prediction in order to 1) characterize locally the city plume on 3 July, and 2) sample the travelling plume in a Lagrangian manner on 4 July. The aircraft quicklook is a longitudinal cross section of approximately 200 km long. Barcelona is overpassed around 1405UT. The vertical structure observed by the airborne lidar is very similar to that observed from the ground. It is worth noting how the multi-layer structure observed above the city vanishes as the airplane moves away from the coast.

A comprehensive model-observation intercomparison exercise based on the 2012 dust IOP is ongoing and involves the NMMB/BSC-Dust model developed at the Barcelona Supercomputing Center (BSC) [5].

Related to the 2012 campaign subject, a potential operationality exercise was performed by 10 EARLINET/ACTRIS and 2 non-EARLINET lidar stations around the northern Mediterranean basin for 72 hours (9 July 0600UT – 12 July 0600UT). EARLINET (European Aerosol Research Lidar Network) is the aerosol vertical profiling infrastructure of the European project ACTRIS (Aerosols, Clouds, and Trace Gases Research Infrastructure). The exercise itself is not linked to ChArMEx activities, but the data are being used by researchers actively involved in ChArMEx for a tentative to assimilate lidar profiles in an air quality model. The same data might also be used in the CALIOPE air quality modelling system [6] developed at BSC to assimilate lidar products, such as the planetary boundary layer height.

4 SPANISH ACTIVITIES WITHIN THE WORKPACKAGE 5: DEPOSITION

Deposition is a crucial parameter of the mass budget

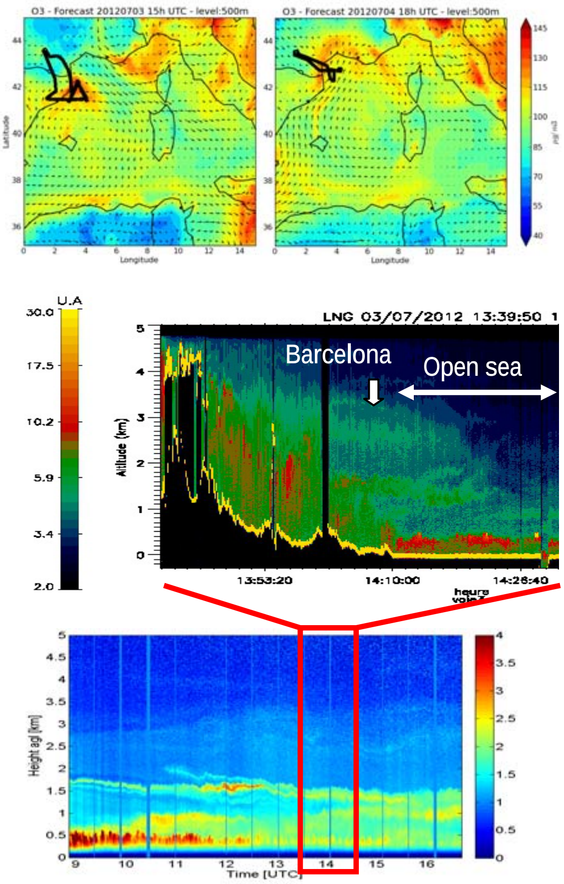

Fig. 5. Pollution recirculation IOP in the area of Barcelona: (top) MACC forecast of O_3 and aircraft track (black lines) for 3 and 4 July; (middle) airborne lidar quicklook on 3 July; (bottom) Barcelona lidar quicklook on 3 July.

of atmospheric particular matter far from their source. In aerosol regional transport models that parameter is not constrained because of the lack of adapted measurements [7][8]. In the marine biogeochemical cycle, the effects of deposition on processes such as phytoplankton bloom and concentration increase of dissolved metals, among others, are not well characterized [9].

To fill those gaps, a deposition network to assess the total insoluble deposition flux of atmospheric particular matter, and especially of Saharan dust, in the Mediterranean region has been set up with a new type of collector. This new collector is an autonomous deposition sampler called CARAGA (Collecteur Automatique de Retombées Atmosphériques à Grande Autonomie). Among the seven instruments currently installed in the western Mediterranean, two of them are in Spain: one in Ses Salines (at sea level and functioning since January 2012), in the Southeast of Mallorca, and one in Cáñar (at 1729 m asl and functioning since August 2012), in the Southwest of Sierra Nevada (see Fig. 1). All systems are performing weekly dust deposition on a network basis. The chemical analyses of the first filters collected are ongoing.

In parallel to this network activity, weekly deposition of wet and dry particulate matter has been collected in Can Llompart since 2010. Fig. 6 shows the inorganic component speciation performed from the deposition measurements.

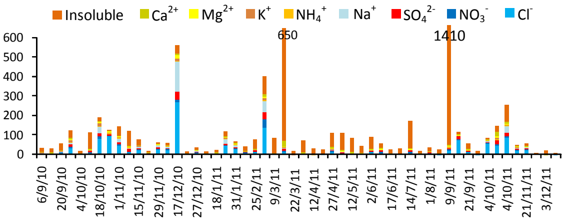

Fig. 6. Inorganic components (in $\mu g \cdot m^{-2}$) of the weekly deposition fluxes collected at Can Llompart.

5 Summer 2013 main field campaign

The 2013 field campaign will take place between 10 June and 10 August 2013. Activities will be mainly in the western Mediterranean. As in the 2012 pre-campaign, the teams will be deployed on a dozen sites including ground measurement stations, balloon launch pad and aircraft bases. Ground operations will be continuous from June 10 to August 10 while airborne operations are organized into two periods of special observations (SOP): SOP-1a from June 12 to July 5 and SOP-1b from July 23 to August 9.

Like in 2012, the campaign is divided into two campaigns defined with different objectives: SOP-1a (ADRIMED, Aerosol Direct Radiative Impact on the regional climate in the MEDiterranean region) will focus on aerosol direct radiative forcing (column closure and regional scale) and SOP-1b (SAFMED, Secondary Aerosol Formation in the Mediterranean) will focus on organic chemistry and secondary aerosol formation.

Both research aircraft involved, the ATR-42 and the Falcon 20, will be based in Corsica, France. In-situ measurements will be performed at several surface stations, including supersites in Corsica, Lampedusa (Italy) and Mallorca. Drifting balloons equipped with an O_3 sonde or an optical particle counter will be launched from Menorca.

As far as Spain is concerned, lidar measurements will be performed in Barcelona and in Cap d'en Font, Menorca, and drifting balloons will be launched from the Sant Lluis aerodrome, Menorca, during SOP-1a; in-situ measurements will be performed in Cap Es Pinar in the north of Mallorca during SOP-1b (see Fig. 1). At the latter site, real time measurements of chemical composition of the gas and particle phases will be performed, as well as offline collection of PM onto filters for subsequent analysis of the organic and inorganic fraction as performed in Can Llompart.

6 CONCLUSIONS

The participation of the Spanish aerosol community either in field campaigns or in meetings and working groups related to the ChArMEx project is remarkable. Spanish researchers are actively involved in the workpackages 1 (emission and source apportionment), 4 (radiative forcing and climate impact) and 5 (deposition) of ChArMEx. Their participation also contributes, less directly, to workpackages 2 (ageing) and 3 (air quality and transport). In particular, the location of the Balearic Islands in the middle of the western Mediterranean Basin where the background conditions are usually clean is a strategic spot for investigating long-range transported aerosol issues such as chemistry, ageing and transport. Overall it makes no doubt that all activities led by Spanish researchers and their international collaborators will yield a better understanding of the Mediterranean aerosols, and an improvement of aerosol chemistry and climate models in the Mediterranean region.

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Table 1. Additional instrumentation installed in Can Llompart during 2010-2012.

	OPC	HV sampler PM10	HV sampler PM1	D/W deposition	Number	MAAP	NH3
Routine	Non stop (1 hour)	Non stop (1 sample/4 day)		Non stop (weekly)			
Sping and summer 2011	Non stop (1 hour)	15/03-15/09 (143 samples)	16/03-21/07 (83 samples)	Non stop (weekly)	15/03-28/04 (44 days)	15/03-15/09 (185 days)	
Summer 2012	Non stop (1 hour)	06/06-15/08 (45 days)	06/06-15/08 (45 days)	Non stop (weekly)	06/06-06/08 (40 days)		06/06-15/08 (50 days)