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Modeling of the chemical composition of fine particulate matter: Development and performance assessment of EASYWRF-Chem

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ABSTRACT

The European emission Adaptation SYstem for the WRF-Chem model (EASYWRF-Chem) has been developed to generate chemical information supporting the WRF-Chem requirements from any emission inventory based on the CORINAIR methodology. Using RADM2 and RACM2 mechanisms, "emission species" are converted into "model species" thanks to the SAPRC methodology for gas phase pollutant and the PM_{10} and $PM_{2.5}$ fractions. Furthermore, by adapting US EPA $PM_{2.5}$ profiles, the processing of aerosol chemical speciation profiles separates the unspeciated $PM_{2.5}$ emission into five chemical families: sulfates, nitrates, elemental carbon, organic aerosol and unspeciated aerosol. The evaluation of the model has been performed by separately comparing model outcomes with (i) meteorological measurements; (ii) NO_2, O_3, PM_{10} and $PM_{2.5}$ mass concentrations from the regional air quality monitoring network; (iii) hourly-resolved data from four field campaign measurements, in winter and in summer, on two sites in the French northern region. In the latter, a High Resolution — Time of Flight — Aerosol Mass Spectrometer (HR-ToF-AMS) provided non-refractory PM_1 concentrations of sulfate, nitrate and ammonium ions as well as organic matter (OM), while an aethalometer provided black carbon (BC) concentrations in the $PM_{2.5}$ fraction.

Meteorological data (temperature, wind, relative humidity) are well simulated for all the time series data except for specific events as wind direction changes or rainfall. For particulate matter, results are presented by considering firstly the total mass concentration of $PM_{2.5}$ and PM_{10}. EASYWRF-Chem simulations overestimated the PM_{10} mass concentrations by +22% and +4% for summer and winter periods respectively, whereas for the finer $PM_{2.5}$ fraction, mass concentrations were overestimated by +20% in summer and underestimated by −13% in winter.

Simulated sulfate concentrations were underestimated and nitrate concentrations were overestimated but hourly variations were well represented. Ammonium particulate matter was well simulated for all seasons. Although simulated particulate OM concentrations in $PM_{2.5}$ were underestimated, their hourly variations were well reproduced by the model. At least BC measurements revealed that EASYWRF-Chem forecast performance was higher in winter than during summer when BC concentrations were very low.

1. Introduction

France, as well as other European countries, has to comply with EU air quality standards (IP/10/1420) but has failed so far to effectively tackle excess concentrations of airborne particles with an aerodynamic diameter less than 10 μm (PM_{10}). In 16 areas in France, the 50 μg m^{-3} daily limit value has been exceeded for more than 35 days per year. Among these 16 areas, the French Nord-Pas de Calais region covers 12,000 km^2 and is populated by 4 million inhabitants with 3 metropolitan areas. Heavy industrialization and a high traffic density contribute to a large amount for NO_X and particulate matter emissions. In this context, air quality modeling can be a useful tool to analyze the origin and fate of pollutants and help enforce emission reduction policies. Therefore, model ability to reproduce past pollution episodes needs to be evaluated in order to make sure that emission scenarios derived from reduction policies are relevant. Moreover, air quality models are useful for atmospheric research since sensitivity studies can help to describe some atmospheric processes. The lack of information about aerosol chemical and physical properties in modeling systems is one of the future research challenges to improve the accuracy and quality of chemical weather forecasting (Baklanov et al., 2014; Kukkonen et al., 2012). Furthermore, improving aerosol modeling is a critical step to enhance emission reduction policies and the knowledge about aerosol impacts over radiative forcing, cloud interaction and human health. In this work, our aim was to build an anthropogenic spatialy-resolved emission

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inventory for the chemical speciation of primary aerosol complying with the WRF-Chem model requirements.

A European Emission Adaptation SYstem for the WRF-Chem code, EASYWRF-Chem, was developed in Python 2.7 in order to adapt European data to WRF-Chem while maintaining the integrity of information. The implementation of aerosol chemical information has been tested, using the same approach as previously initiated by the UPM (Universidad Politecnica de Madrid) and the UCM (Universidad Complutense de Madrid) (Borge et al., 2008; San José et al., 2010). Emissions of the elemental carbon and primary organic fractions of aerosol were sometimes not treated or simulated using specific emission inventories (Bessagnet et al., 2008; Schaap et al., 2004). On the contrary, Borge et al. (Borge et al., 2008) has presented an adaptation of the Sparse Matrix Operating Kernel Emissions model (SMOKE) (Houyoux et al., 2004) to generate a Spanish emission inventory based on the CORINAIR methodology. Similarly, San José et al. used the European chemical speciation data. Studies of Borge et al. and San José et al. have highlighted challenges to overcome for transferring European data to the US Environmental Protection Agency (EPA) database and chemical speciation system without corrupting data integrity. Our work has combined both architectures by using the European gaseous chemical speciation and the US EPA particle chemical speciation to implement a new methodology based only on the EMEP emission inventory. The simulation results were statistically compared to (i) ground-based measurements from the Météo France French weather network; (ii) pollutant concentrations measurements for ozone, nitrogen dioxide and particulate matter (PM_{10} and $PM_{2.5}$) from the French air quality monitoring network ATMO; and (iii) field measurements of fine particles chemical composition from a High Resolution — Time of Flight — Aerosol Mass Spectrometer (HR-ToF-AMS) and an aethalometer.

2. Methodology

2.1. Ground-based atmospheric measurements

EASYWRF-Chem simulations were compared to ground-based measurements over the French northern region. Meteorology modeling has been statistically evaluated by comparing the results to hourly-resolved measurements from 16 stations of the Météo France network (Fig. 1). The following parameters have been used: temperature at 2 m (T), relative humidity (RH), rainfall (RF), wind speed (WS) and wind directions at 10 m (WD).

Concerning pollutants, NO_2, O_3, $PM_{2.5}$ and PM_{10} hourly-resolved concentrations were gathered from 22 stations of the ATMO-Nord-Pas de Calais air quality monitoring network reported in Fig. 1. Location and station types are detailed in appendix A.

Particle composition measurements have been performed at two sites during two separate periods of about 30-days in winter and summer seasons (Table 1). The Dunkirk sample site is an urban background site influenced by the industrial port of Dunkirk, with petroleum processing and steel production industries and presented in Fig. 2. Moreover, the proximity of the site to the North Sea makes air quality modeling challenging due to specific atmospheric dynamics such as sea breeze phenomena especially during summer (Boyouk et al., 2011; Roukos et al., 2011).

The second site was located in Douai, a typical urban background site surrounded almost exclusively by residential areas and close to a major highway system at ~2 km. Douai is also surrounded by agricultural areas.

A High Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS), described in many previous publications (e.g. (Aiken et al., 2009; DeCarlo et al., 2006)), was deployed at the two sampling sites during both seasons. It was proven useful to measure the real-time

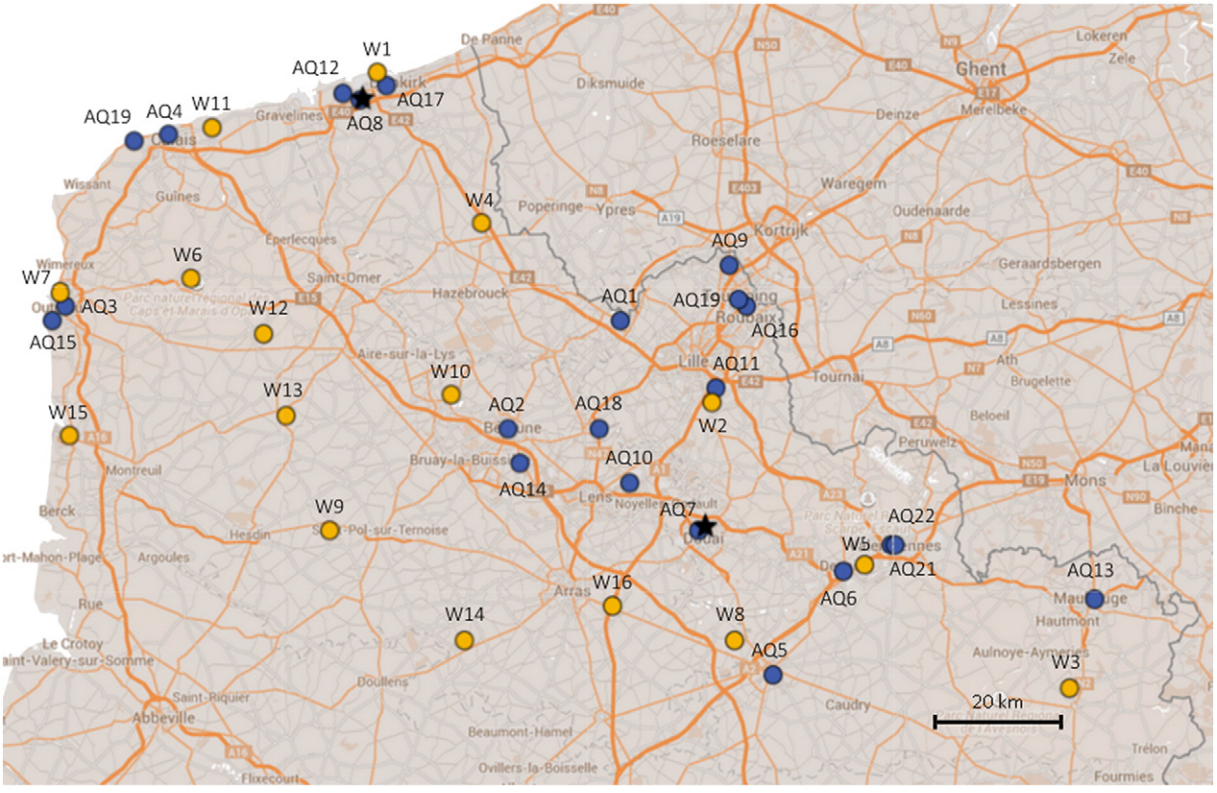

Fig. 1. Map of weather stations (yellow dots), air quality monitoring stations (blue dots) and HR-ToF-AMS measurement sites (black stars) over the French Northern Region. Weather and Air Quality stations respectively named W1 to W16 and AQ1 to AQ22 are presented in Appendix A.