

# Regionally Significant Residential-heating Source of Organic Aerosols

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## Abstract

Aerosols play an important role in the extreme events of air pollution and climate change. Despite the decade's efforts, air pollution models are still unable to simulate the heavy pollution events very well due to the complicated aerosol chemical composition, size distribution and spatial and temporal variations. Recently, the aerosol mass spectrometer (AMS) was deployed in Dublin to determine the chemical composition and concentration of submicron aerosols at high temporal resolution. The organic aerosol (OA) was found to dominate the aerosol composition (>50 %) in the heavy pollution events. The particular matter concentration (PM<sub>1</sub>) was measured to exceed 200 µg m<sup>-3</sup> on 19 November 2016 and 22 January 2017.

In this study, we use the WRF-Chem model to simulate the heavy organic aerosol episodes in Dublin. However, the model could not rebuild the heavy pollution events with current emission datasets (EDGAR, EMEP, TNO, NEI or RETRO) we use. To have better simulation results, we have examined the sensitivities of simulation results to the emissions, meteorological parameters or spatial and temporal resolutions. We find that the enhancement of local emissions with the reference of measurements including diurnal variation and temperature dependence is the most effective way to rebuild the heavy pollution events. The heavy organic aerosol episodes in Dublin are found to be contributed mostly from local emission sources (peat burning). The lack of accurate speciation and temporal profiles in current emission datasets should be greatly concerned, particularly the significant underestimation of organic carbon (OC) emissions. The uncertainties in current emission datasets will hamper us to forecast the heavy air pollution events accurately and improve the early warning system from forecasts, hence it is imperative to improve the emission inventories using the new method of integrating top-down and bottom-up approaches with available measurements.

## Introduction

The tiny particles (particulate matter or aerosols) play an important role in the air pollution and climate change. It can either reduce the visibility and be harmful to the human health or scatter and absorb the sunlight which affects the radiation budget directly and affects the climate. It can also act as Cloud Condensation Nuclei (CCN) to affect the cloud formation and affect the climate indirectly [1,2]. Despite much progress has been achieved in recent decades, the aerosol burden and forcing remain uncertain due to the complicated aerosol chemical composition, particle shape, size distribution and temporal and spatial distributions. The indirect effect of aerosols is much more puzzle. Indeed, aerosol is one of most uncertain components for evaluating the air quality and climate effect and predicting future air quality and climate change. The aerosol species (sulphate, nitrate, ammonium, black carbon, organics, dust and sea salt) have been identified and simulated in most regional and global models, most aerosol species have been simulated well however, the organics, dust and sea salt simulations remain uncertain, particularly the primary and secondary organic matter [3].

In China, the residential coal burning has been considered as the important source of organic aerosol [4,5]. Recently, the aerosol chemical speciation monitor (ACSM) was deployed to study the primary non-refractory submicron particulate matter emissions from the burning of commercially available solid fuels (peat, coal, and wood) typically used in European domestic fuel stoves. The resolved oxygenated organic aerosol (OOA) dominated the aerosol composition in continental air masses, with contributions of 50%, compared to 12% in marine air masses [6]. The highest measured concentration in Dublin of PM<sub>1</sub> can reach up to 250 µg/m<sup>3</sup> on 22 January 2017 and 19 November 2016. These organic aerosol episodes occurred in Dublin during winter season, particularly during the coldest days. The local

emission sources have been found to be the residential heating, and most organic aerosol emissions are contributed from the peat burning [7].

From satellite observation, regional hotspots of carbonaceous aerosols from biomass burning, fossil fuel and biofuel combustion have been identified in east Asia, south Asia, central Africa, western North America, South America and Europe [8,9]. Hence, it is of great interest to examine the carbonaceous aerosols simulations from global and regional air quality models by space-based or ground-based measurements. The Air Quality Model Evaluation International Initiative (AQMEII) focuses on regional modeling domains over Europe and North America [10], within which aerosol meteorology interactions were studied [11,12]. Online coupled models have been used to study the meteorology and chemistry interactions, but many important questions remain [13,14]. Evidence is accumulating that emissions of carbonaceous aerosols from residential solid fuel burning in the UK have been underestimated and/or spatially misclassified [15].

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To reproduce the aerosol episodes using global or regional model is a challenging task. The high concentrations of air pollutants are not only contributed from the local sources, but also contributed from the long-range transport. The simulations of aerosol concentrations depend on the emissions, meteorology parameters, physical and chemical processes. In this study, we use the WRF-Chem model to simulate the Dublin heavy pollution events.

## Description of WRF-Chem Model

The WRF-Chem model is the Weather Forecasting Research (WRF) model fully coupled with chemistry [16]. The model simulates the emission, transport, mixing, and chemical transformation of trace gases and aerosols simultaneously with the meteorology. Gas-phase chemistry and aerosol schemes are integrated over the same time step as transport processes, allowing fully coupling between schemes and making sensitivity studies on the feedbacks between meteorology and chemistry possible.

## Model configuration

WRF-Chem Version 3.9 is used for model simulations of Dublin air pollution events. The centre of domain is located at 52.32N, 1.51W with resolution 25 km, which covers Ireland, UK and most European regions. The vertical structure is divided into 30 levels, of which 8

levels are lower than 1 km high. The terrain, land use and soil data are interpolated into model grids from the USGS global elevation with resolutions of 10 m, 2 m and 30 s, respectively. The gas-phase chemical mechanisms used here are Regional Acid Deposition Model version 2 (RADM2) [17] and organic gas-phase chemical mechanism CRIMECH v2 [18]. The aerosol scheme is the Modal Aerosol Dynamics Model for Europe (MADE/SORGAM) [19,20]. The MADE scheme has 3 modes (Aitken, Accumulation and Coarse), which can be used to investigate the direct and indirect effects of aerosols.

## Model experiments

The simulation period ran from January 20<sup>th</sup> to January 24<sup>th</sup>, 2017, covering one of the most severe PM<sub>1</sub> pollution episodes that occurred in January 2017. The other simulation period ran from 17<sup>th</sup> November to 24<sup>th</sup> November 2016, covering the severe PM<sub>1</sub> pollution episode that occurred in November 2016. The ECMWF ERA-Interim data files are used for the meteorological initialization and boundary conditions. We use the TNO and NAEI emission datasets to produce the emission input files for the WRF-Chem run. Aside from the PRE-CHEM emission processor, we have another emission pre-processor from Manchester University, which can be used for processing the hourly emission files for WRF-Chem run, particularly for the simulation of heavy pollution events. The ECMWF CAM forecast datasets have been used for the chemistry Boundary Conditions (BCs).

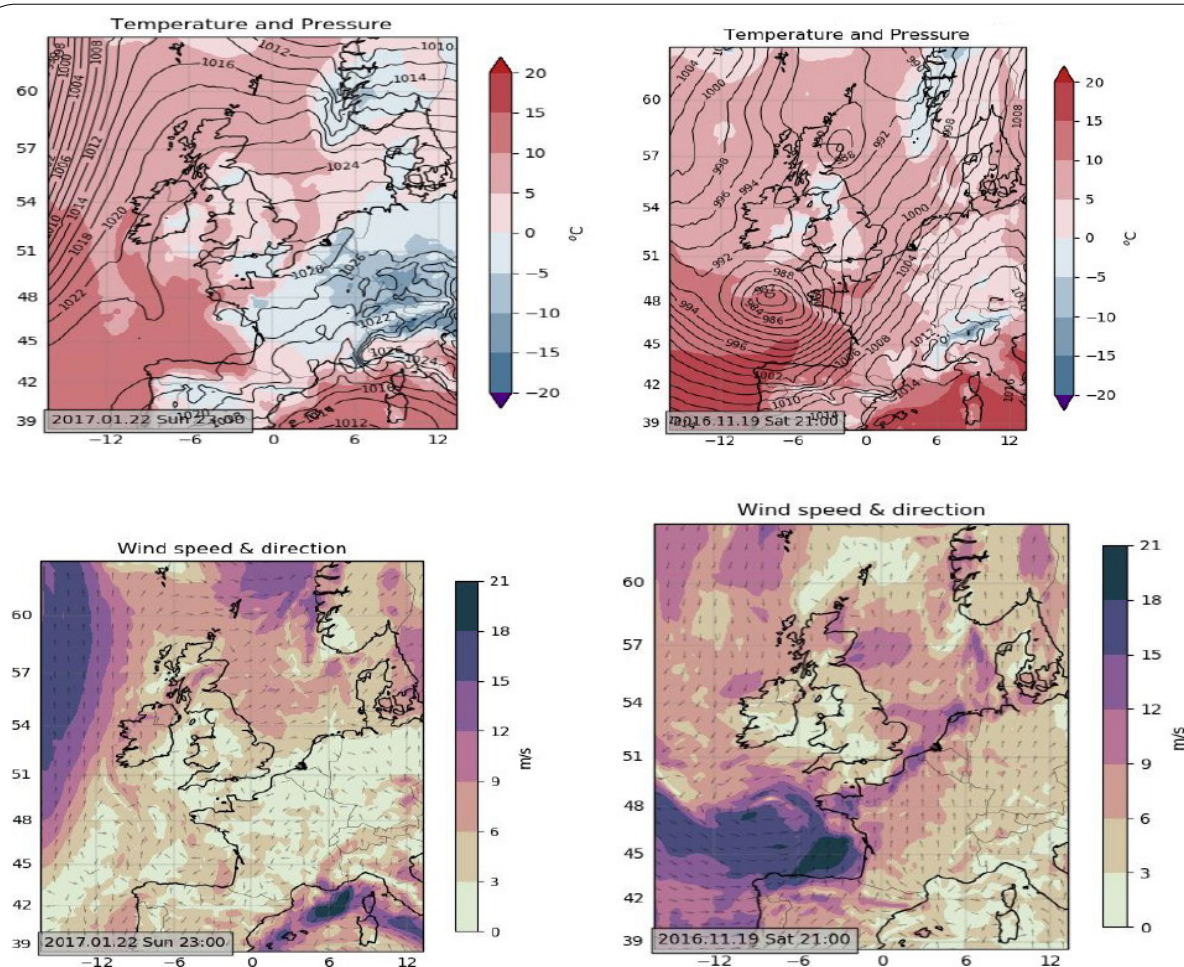


Figure 1: The left side: temperature, pressure and wind fields on 22 January 2017. The right side: temperature, pressure and wind fields on 19 November 2016.

## Results

### Meteorology

On 22 January and 23 January 2017, low temperature and high-pressure system dominated most part of Europe including UK and Ireland (Figure 1). The specific humidity is high and southerly wind with low wind speed in Dublin during the heavy pollution episodes (Figure 1). On 19 November and 20 November 2016, the low temperature and low wind speed occurred in the Dublin region (Figure 1). The meteorology conditions were favourable for accumulating air pollutants in that period.

Figure 2 and 3 present the inter-comparison of WRF-Chem model results with measurements at Dublin Airport. The measurement data

were collected from Met Eireann's weather observation network. We find the model simulations of wind speed and surface temperature have overall good agreement with the measurements although there is a discrepancy between model simulations and measurements sometimes. We have performed the sensitivity of model simulations to the horizontal resolutions. The model simulations with high resolution (1 km) have the better results.

### PBL schemes

WRF-Chem functions well in replicating the meteorology during these events. We have performed sensitivity experiments to investigate the ability of different boundary layer parameterisation to simulate the meteorological conditions that cause pollution events. Figure 4 and 5 show the inter-comparison results of simulated PBL height with

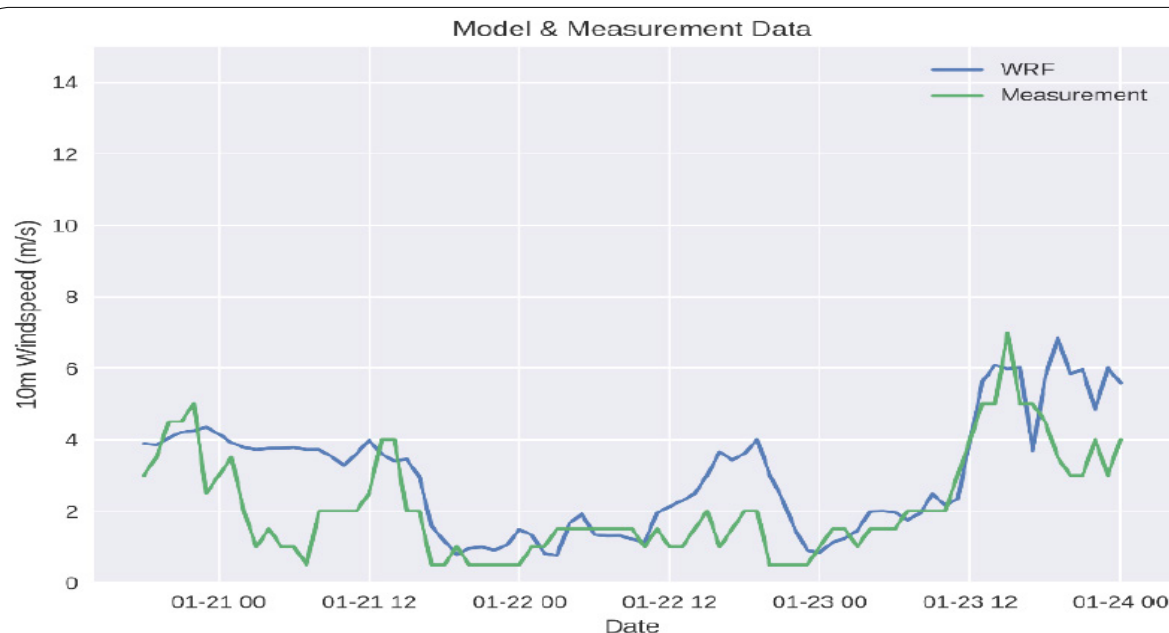


Figure 2: The inter-comparison of wind speed simulated by WRF-Chem and measurements.

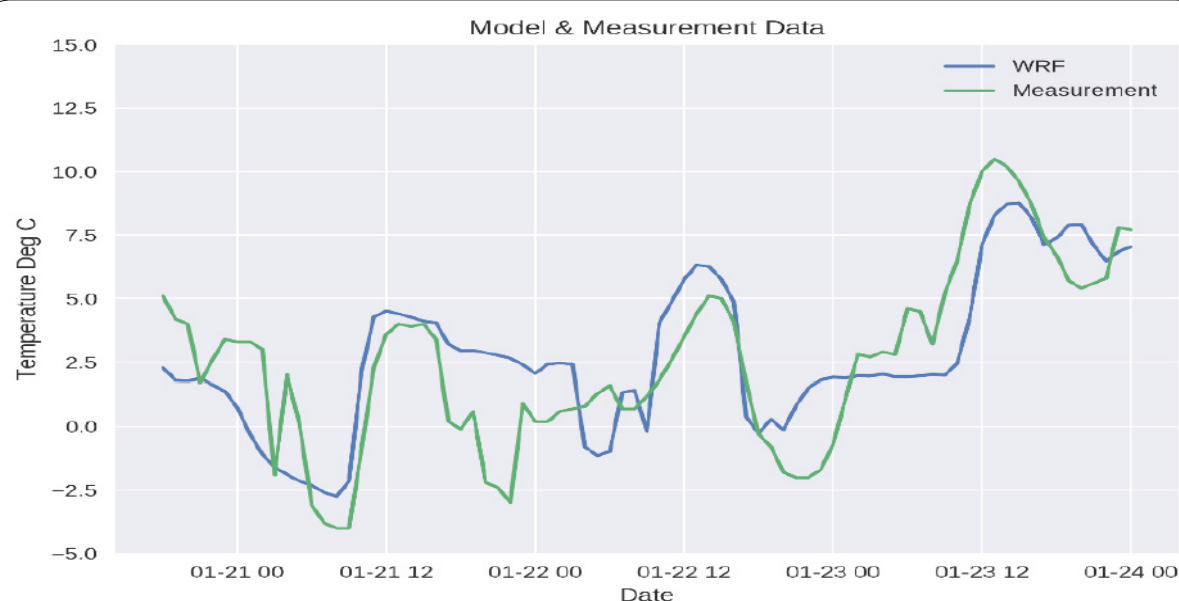


Figure 3: The inter-comparison of surface temperature simulated by WRF-Chem and measurements.



measurements. The measurement data were collected from ceilometer measurements. The YSU boundary layer mechanism functions best in agreement with the boundary layer measurements taken at Casement aerodrome from Met Eireann's ceilometer.

## Emissions

We use the TNO and NAEI emission datasets to produce the emission input files for the WRF-Chem run. We find the problem of emission datasets caused the failure of recreating the pollution events. Currently available highest resolution spatial emissions for Ireland are the MapEire emissions. Emissions of PM<sub>2.5</sub> over Dublin are given as

0.18 Tonnes per year. This equates to  $360 \mu\text{g m}^{-3}$  over a year in the relevant  $1\text{km} \times 1\text{km}$  spatial area (assuming a well-mixed boundary layer of 500 m). Considering one household burning 1kg of wood would emit over 4 million  $\mu\text{g PM}_{2.5}$ , it is extremely likely that the MapEire emissions of  $360 \mu\text{g m}^{-3}$  is an underestimation. This can be attributed to the fact that many people use solid fuel combustion as a secondary heat source in the winter months and do not declare such on census forms. It is proposed that further work be carried out to determine comprehensive temporal and spatial emissions for the burning of solid fuel, without which the forecasting of air pollution events caused by domestic fuel combustion is impossible.

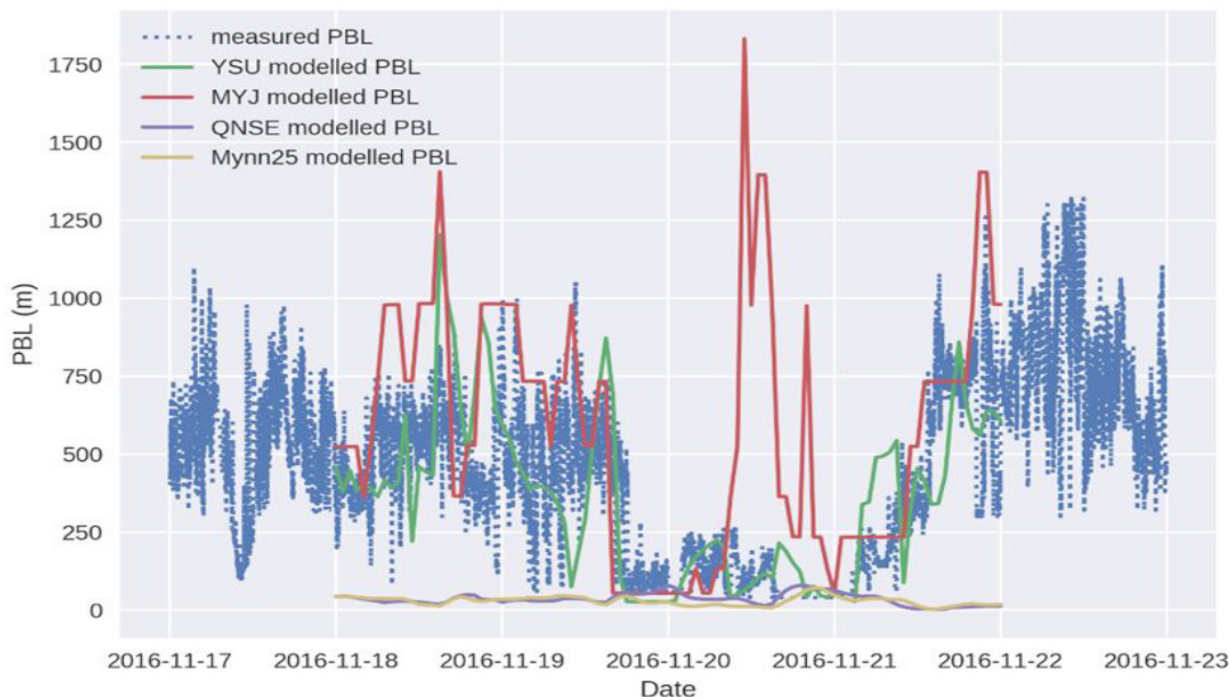


Figure 4: Boundary Layer Height measured and modelled during pollution event in Dublin, November 2016.

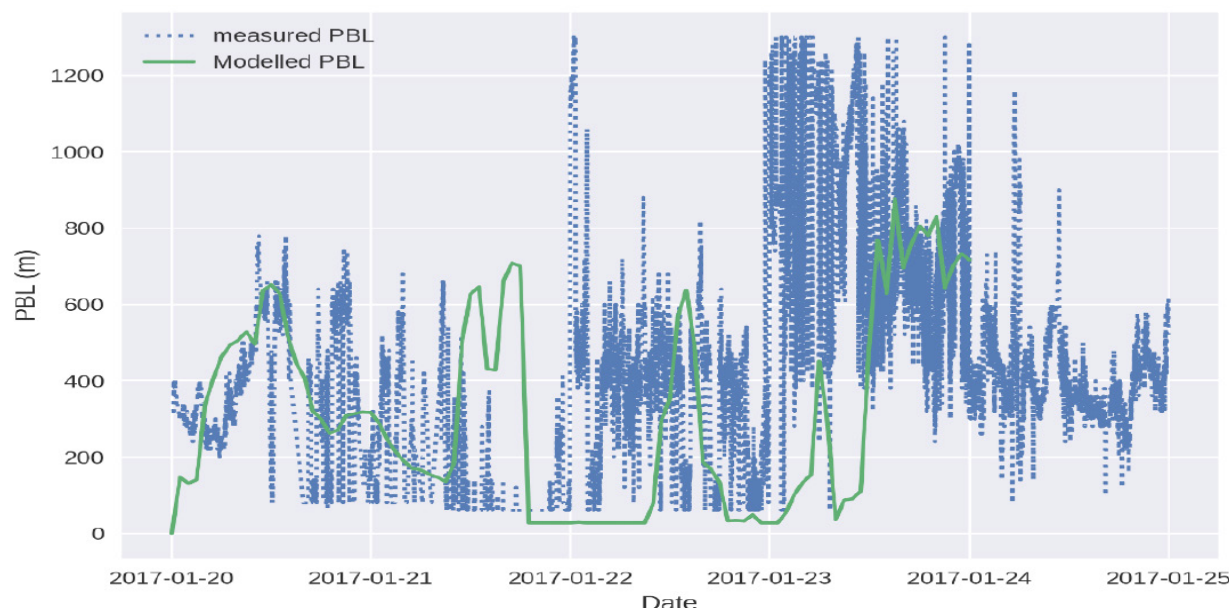


Figure 5: Boundary Layer Height measured and modelled during pollution event in Dublin, January 2017.

## Chemistry schemes

WRF-Chem at C-CAPS was updated to run with advanced detailed organic gas-phase chemical mechanism CRIMECH v2 [18,21], a mechanism that handles atmospheric chemistry more explicitly, especially pertinent for O<sub>3</sub> and VOCs. For simulation of pollution events that have been observed in cities in Ireland, a new formulation for emissions was developed whereby emissions are temporally resolved to correspond with the consumption of solid fuel as a secondary heating source in residential sector.

We have enhanced the local emissions with diurnal variation and temperature dependence. Figure 6 and 7 present the inter-comparison of WRF-Chem simulations with measurements. We can find that the

enhanced local emissions from solid fuel burning have improved the model results and a better agreement with measurements. The core episodes of Dublin air pollution events have been simulated better than the results with TNO and NAEI emission inventory.

## Discussions

Our results show that current emission inventory including EMEP, TNO and NAEI largely underestimate the organic aerosol emissions from residential solid fuel burning. If we use the current emission inventory, we could not reproduce the heavy pollution events. The underestimation of organic aerosol emission appears also in climate modelling. If we underestimate the organic aerosols in climate modelling, it means that our current climate modelling has

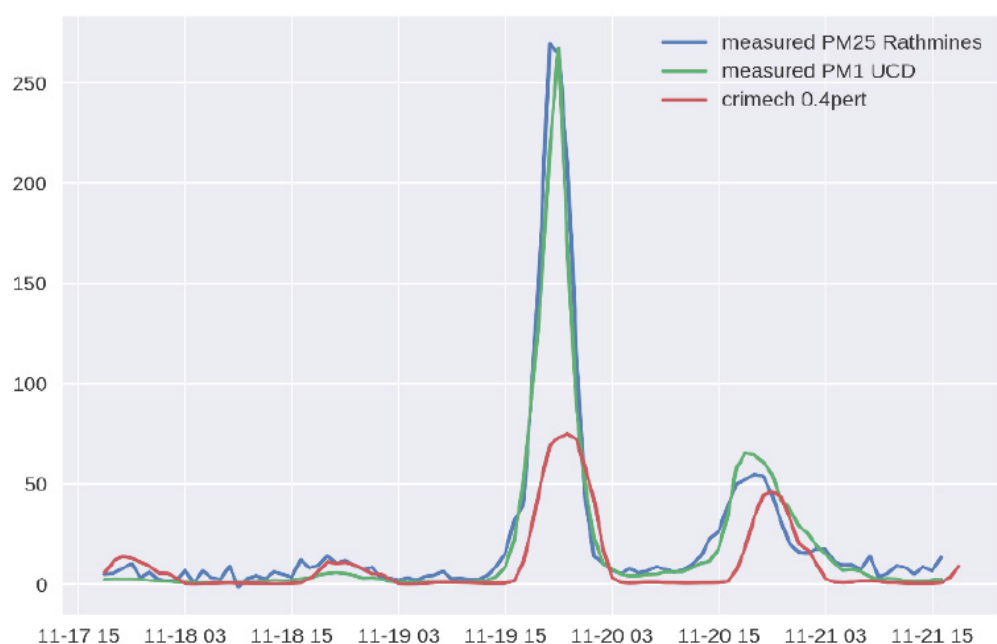


Figure 6: Inter-comparison of PM1 simulations with measurements for November 2016.

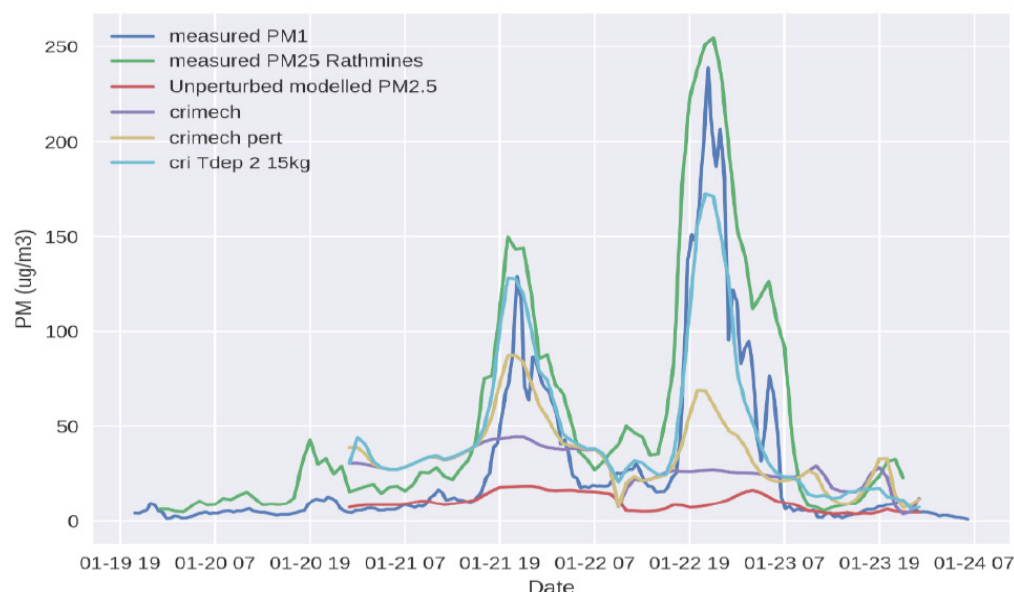


Figure 7: Inter-comparison of PM1 simulations with measurements for January 2017.

underestimated the aerosol negative radiative forcing and the aerosol cooling effect. This could partly explain the recent global warming hiatus (2000-2015) as the climate models underestimate the cooling effect and overestimate the warming effect.

For air quality and climate modelling, the importance of emission input data had been addressed decades ago. Despite the progress in the improvement of emission inventory, the accuracy of emission data used by modelling is still limited in terms of spatial resolution, temporal profiles and speciation. Emission becomes one of biggest uncertainties in model simulations and it is imperative and necessary to continuously improve the emission data. As we know, most emission data sets are created by top-down approach. In recent years, the bottom-up approach has been used widely to improve the emission data. We advocate to use a new method of integrating top-down and bottom-up approaches with available measurements.

## Summary

In this study, we have investigated and simulated the Dublin heavy pollution events using WRF-Chem model. The heavy pollution events are caused by the enhanced local emissions from the residential solid fuel burning, particularly the peat burning during the cold days in winter. The emission data from EMEP, TNO or NAEI underestimated the residential emissions, and led to the large underestimation of organic aerosols. The sensitivities of simulated PM episodes to PBL height, chemistry schemes and emissions have also been studied. With the enhanced the residential emissions during the cold winter, we can reproduce the heavy pollution events in Dublin. Considering our findings on aerosol episodes studies, the underestimation of organic aerosols in the current emission data used by regional and global modelling of air quality and climate is one of most uncertainties in the predictions of future air quality and climate change. More efforts from multiscale modelling and measurements are necessary and envisaged to improve our model prediction accuracy of future air quality and climate change.

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## Competing Interests

The authors declare that they have no competing interests.

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