PUBLISHABLE SUMMARY. AIRPURE. IOF N°: 234961. Fellow: Richard Ramaroson.

General introduction

The EC-FP7 Workprogramme "People" includes the "International Outgoing Fellowships for Career Development (IOF)" aiming to reinforce the career of European individual researchers by ***training and acquiring new knowledge*** in a third country at a ***high-level research organisation***. Project research fields are chosen by the applicant in a 'bottom-up' manner with a ***coherent research programme*** in a view to strengthen, improve and/or diversify his expertise. The IOF full period is divided into: 2-year outgoing phase at Harvard University, USA and 1-year return phase at The French Aerospace Lab., FR.

Project Goal

The project research field N° PIOF-234961 namely AIRPURE aims at investigating air pollution by PM2.5 (particulate matter less than 2.5um) and its speciation and ozone and its precursors at mega-cities, particular focus for Paris mega-city (26th number of population) , after acquiring scientific hand-on experiences and being trained at the outgoing organisation.

Motivation

It is worldwide recognized by epidemiologically (and toxicologically) studies that a robust relationship exists between exposure of population to ambient particulate matters with diameter less than 2.5 µm (PM2.5) and adverse health impacts (lung cancer, heart disease, mortality, etc.). A new European Union Directive 2008/50/EC includes new regulations for ambient PM2.5, a binding cap for annual mean at 25 µg/m^3 coming in force in 2015 whereas in 2006, the US National Ambient Air Quality Standard (NAAQS) was tightened to 15 µg/m^3 on a 24-hour average. PM.2.5 are formed and composed by hazardous air pollutants (HAP), elemental carbon, organic carbon, inorganic species, metals and other species. This project brings a contribution on improving scientific knowledge on PM2.5 and ozone pollution characterisation.

Objectives

The project objectives consist in acquiring/re-enforcing skills and know-how of the fellow to characterize aerosols, particularly PM2.5, and ozone at different scales including the urban mega-cities (US, Europe) by adapting and providing methodologies and holistic tools. In the Description of Work (DoW), the scientific research focuses on three topics: exploring the feasibility to infer PM2.5 map at the ground from satellite Aerosol Optical Depths (AOD), analyzing ground air quality observations; modelling the formation and fate of PM2.5 and ozone and their precursors by adapting and using existing multi-scale emission, weather forecast, and air quality models; and a quality management of the holistic tools composed by the observations, the model data input and the numerical model results. Training activities foreseen in the programme include: satellite data, emission, air quality modelling tools and academic formation on Quantum Cascade Laser (QCL) and their application on environmental sciences. Lectures given by the fellow to Harvard graduate Students have been foreseen.

General Summary and context

The project has been fulfilled at the School of Engineering and Applied Sciences (SEAS, Harvard University): **1)** as central location within the Atmospheric Chemistry Modelling Group (head: Prof. Daniel Jacob)/ACMG, dealing with global modelling, closely and regularly with: **2)** the Environmental Chemistry Group/ECG (head: Prof. Scott Martin) related to laboratory measurements of PM speciation , **3)** the Atmospheric and Environmental Science Group/AESG (head: Prof Steve Wofsy) related to field measurements at the ground using laser techniques; **4)** the Harvard School of Public Health/HSPH (Head: Prof. Petros Koutrakis) for PM2.5 inferring from satellite observations of aerosol and ground monitoring/analyses of PM2.5 speciation in relation to health issues; and additionally with **5)** Prof Frederic Capasso/FC/Harvard University for academic lecture on QCL techniques. Remote scientific cooperation were established with: **6)** the NCAR (National Center for Atmospheric Research, Boulder, CO) for weather research forecasting (WRF model), biogenic-biomass burning emission modelling and databases; **7)** the NASA (National Aeronautics and Space Administration) for AOD retrieval from satellite (MODIS) and databases at high resolution; **8)** the University of North Caroline/UNC for multi-scale air quality models, 9) the US-EPA (Environmental Protection Agency) for anthropogenic emission models and databases and 10) the AERODYNE Research Inc. (Boston) for measurement capabilities using quantum cascade laser for atmospheric applications. **9)** In Europe, much closer cooperation has been started in mid-2010 for emission processing with the Institute HZG (Germany), in 2011 with European research groups dealing with weather forecast for Europe, with AIRPARIF for ground monitoring of PM2.5 and finally using previous ONERA projects outcome on air quality at airports (PM2.5 incl.) for which the fellow has been the Principal investigator and project manager.

Trainings, lectures, outreach, impacts (list): *1) High-level lectures* given by Prof. Frederic Capasso to graduate students on ***QCL theory***, technology and applications during 6 academic months, have been followed fully by the fellow (2010), as new knowledge by strengthening his know-how on innovative laser-based measurement techniques. The lecture demonstrates the QCL powerful capabilities for a broad range of applications namely for this IOF project i.e.: for most of atmospheric pollutants and from industries, for greenhouse gases, spanning the near-, mid-, and far-infrared spectra applications. **2)** The lecture has been completed by regular ***scientific cooperation*** with AESG for ***measurement of atmospheric species using QCL*** (e.g. laboratory and field observations at Harvard Forest) and by a technical visit of AERODYNE Inc. laboratories: a QCL-based instruments developer. Other QCL applications encompass powerful instruments dealing with military applications (engines, gas detection, and counter-measures). QCL-based instruments are widely used internationally for atmospheric applications but very limited in Europe compared to US despite their strengths, high-quality, high frequency, high power and large spectral spanning band. **3)** Inside SEAS, regular participation to ECG group meeting and scientific cooperation with its researchers ensured comprehensive and better understanding of ***PM2.5 chemical composition (using Aerosol Mass Spectrometry)*** and their pertinent role on AOD retrievals from satellites. **4)** One-week ***training on air quality modelling tools*** organized by UNC has been followed by the fellow strengthening and deepening his knowledge of the Community Multiscale Air Quality model (CMAQ). **5)** EPA and UNC provided one-week ***training on emission modelling and databases*** for which the fellow applied as well. Unlike in Europe where emission databases start to be elaborated sparsely, EPA has developed for the community, comprehensive models and databases for anthropogenic emissions creation (multiscale) using detailed information from all sectors of emission in US and required as input by multi-scale air quality (AQ) models. However, adapting these tools for European applications requires a certain amount of man powers and was not proposed in this project. **6)** Due to non-existence of appropriate emission databases for multiscale air quality modelling in Europe, ***cooperation*** with the Institute HZG in Germany has been initiated to ***generate European multiscale anthropogenic emission databases*** used as input by AQ model applied to European regions and by adapting the SMOKE EPA model. This close cooperation is successful and leads to a first set of emissions data for summer conditions. The existing European EMEP emission database is not appropriate for multiscale air quality modelling. This work constitutes an extra task in the project requiring more person-months, and not foreseen, but scientifically relevant to achieve the project objectives; the fellow decided to perform the task. **7)** The last training deals with ***satellites NASA A-Train*** (e.g. MODIS, OMI, CALIPSO) for AQ studies, analyses and retrieval/use of AOD and other air pollutants (AP). The lecture (2010) has been a high-level added-value to the fellow expertise and strengthen his knowledge in better characterising AP (gas, aerosols) observed from different satellite platforms along with ground observations, necessary to improve his capabilities on analyzing observations versus AQ model results. **8)** In the other side, ***the fellow gave 2 lectures*** to graduate/PhD Students at ACMG and at HSPH on air pollution measurement analyses at airports and on air pollution modelling. **9)** The fellow has participated to a one day training of impact-valorisation of nanotechnology (including nano particles) followed by one-day ***outreach for the general public*** at Boston Museum of Science organized by Harvard University team. **10)** The fellow attended (**as training**) ***Workshops***/Conferences regularly organized by the University of Davis (US, CA) dealing with Atmospheric Aerosol Algorithms and Modelling and aerosols speciation particularly focused on the ***role and importance of organic aerosols*** on atmospheric particulate matters. These workshops gave him opportunities to improve his knowledge of organic aerosol:, their volatility and origins, their contribution to PM2.5 formation as secondary organic aerosols as well as from both sources: anthropogenic and biogenic.

Summary of Scientific Research, work and results

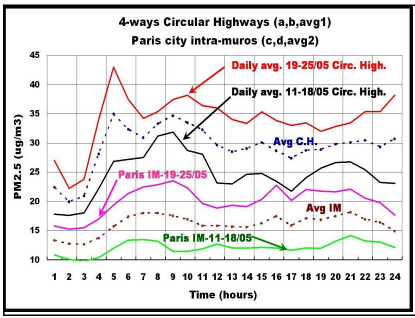
As described in detail in the mid-term and periodic report, the work plan includes **two phases**: **1)** acquiring, strengthening, improving know-how and scientific expertise, adapting tools (data, measurements, model results) to US air quality studies and **2)** adapting, applying and implementing the holistic tool to European countries with a particular focus on Paris mega-city. The main body and skeleton of the scientific project reside connecting and merging: ***observation-measurement data, emission source databases, weather forecast output and AQ model results*** from continental to local scale to constitute a holistic tool for air quality studies and prediction.

a) For emission processing and database creation (AQ model input) for both phases (which is not finally only the compilation as proposed in the DoW), the EPA SMOKE emission model has been adapted to all domains for US and Europe. Both studies are applied to 4 nested domains respectively: at 45km grid domain resolution for the biggest domain 1 (continental US and continental Europe), at 9km resolution for regional domain 2, at 3km resolution for regional-urban domain 3 (including Ile de France for Europe and New England for US), and at 1km res. for local scale centered on Paris and on Boston respectively. ***Anthropogenic sources:*** The SMOKE model has been successfully adapted and run for EU regions and US respective domains. For US, the huge and detailed amount of input data required by SMOKE is available to the scientific community. In Europe, such databases are not yet comprehensive enough but sufficient to create gridded emission data for EU domains in this project. This is an innovative approach thanks to a close cooperation with HZG. Anthropogenic emissions are not only the source of air pollutants (AP). ***Biogenic sources*** emitted by forests, lands (wet, dry) contribute significantly to emissions of reactive volatile organic compounds (VOC) precursors of ozone and forming Organic Compounds (OC) into aerosols. Unlike in US, such emission databases for multiscale modelling do not exist in Europe. The NCAR MEGAN model dedicated to calculate speciated emissions of biogenic VOC are adapted on multiscale approach, using land cover, vegetation types, leaf area index retrieved from MODIS satellite observations at 1km res. and by using hourly meteorological variables (as emissions depend on temperature and solar radiation) outputted from the weather forecast model performed in this project. Biogenic emissions are mapped for the period of modelling studies, separately for each domain, in US and in Europe i.e. for all air quality scales. These emissions are among unique ones in Europe. ***Biomass burning emissions***: Biomass burnings emit multiple AP including organic compounds, CO, CO2, smoke, and other species. MODIS observations allow the discrimination of burned areas at 250m resolution based on surface pixel temperature. The NCAR FINN emission model dedicated to generate biomass burning emissions (speciated) is adapted to multiscale approach for Europe and US domains on a daily average basis using MODIS data and redistributed on hourly-frequency basis. Emission factors of pollutants are collected from literature. Multiple scales emission databases are created for European domains studied in this project which are unique and innovative as outcome of the project. They may be used as AQ model input or for any future abatement strategies in Europe. ***b) Weather forecast and meteorology (WRF):*** Air pollution dispersion is highly meteorologically-dependent. Depending on AQ modelling study objectives, the weather fields must be adequate accordingly, i.e. ***"the AQ scales must be consistent with the scales than the weather forecast model can resolve".*** In other words, ***using global meteorological fields at low resolution is a wrong approach to model air pollution dispersion at local scale***. Herein, the AQ model solves time-dependent air AP evolution onto nested 3-dimensional domains for different AQ scales: local, urban, regional, and continental. As the weather and air pollution are decoupled (model approach), the WRF model has been adapted and run to provide hourly-based meteorological forecast over each period of investigations; domains are interacting to ensure interaction of scales. WRF requires a huge amount of observed data: collected, analyzed and merged from ground stations, radio sondes, satellites, aircraft, and ships. These 3D and time-dependent databases are extracted from NCAR storage systems and post-treated in this project. ***c) AQ modelling:*** The central modelling task performed in this project is the AQ prediction of all air pollutants i.e.: adaptation of CMAQ to US and EU domain modelling (adaptation and integration of hourly 3D emissions input and meteo fields), integration of new routines (use of more performing Ramaroson's numerical scheme for the chemistry), improvement, implementation, run, post-treatment, exploitation of numerical results, deep analyses versus some observations (ground and satellites) and adaptation of boundary conditions from the ACMG/GEOS-CHEM global model. Model runs have been performed using two different system computer architectures: US computers at Harvard University and the French Aerospace Lab. high performance computers. Incompatibilities between CMAQ and Harvard system in the other hand, and between US systems vs. French's have been source of multiple issues for model implementation and create delays. Despite these non-negligible problems, air pollution modelling studies using CMAQ have been performed successfully for both objectives: US & EU and for all scales herein including some mega-cities (New York, Boston, and Paris). CMAQ solves the 3D time-evolution of AP, particulate matters and their chemical composition/size distribution at different AQ scales for all domains. Models results (US&EU) have been stored, after post-treatment and analyses on an hourly and daily basis in order to compare to available observations. ***d) Observations:*** The collect/analyses of air pollutants observations at the ground and the MODIS satellite AODs used to infer PM2.5 ground distribution map for the periods of investigations, constitute the second main components of the project. **AQ ground observations**: For the US AQ studies, EPA's Air quality system (AQS) consists of 947 filter-based daily and 591 continuous stations regularly updated on its web site until now, focuses on urban area such as mega cities measuring multiple AP including PM2.5. PM2.5 chemical composition is monitored by a limited number of specific non EPA AQ sites such the Harvard SPH supersite in Boston. Rural and regional AP trends are monitored by specific stations networks. Ground data corresponding to the periods of interest and spanning over the biggest domain 1 have been collected, analyzed vs. models and meteo fields along with the HSPH supersite AQ data in Boston. In Europe, "regular" ground observations and monitoring of AP are collected by AIRBASE (raw data) and EMEP (for model use). All EU AQ stations do not include necessarily major AP; some stations observe daily rather than hourly. In addition, these data have been stored only until 2007. PM2.5 is not monitored by all Member States (e.g. not by France except AIRPARIF-Paris), unlike in US. Some limited-in-time field campaigns include PM10 and PM2.5 speciation and limited in number of observation locations. However, some limited-in-time measurement campaigns organized by European projects such as MEGAPOLI and EUCAARI provide PM2.5 speciation (e.g. organic compounds, sulphate, nitrate, ammonium, etc.) very useful for model evaluations. AIRPARIF, France (www.airparif.asso.fr) is for instance the only French provider, to the public, of PM2.5 (along with other AP) via networks of monitoring stations at Paris mega cities and its surroundings from urban to rural and along highways. EMEP and AIRPARIF databases have been used to extract AP concentration (with PM2.5) for the modelling case studies for Europe. **As an application of the holistic tool,** the periods of model investigations and evaluation against observations have been chosen with regards to pollution events observed from MODIS satellites and from ground AQ stations. For US, it spans from August 15th to August 30th 2007 during which multiple hot spot pollution events have been observed for North-East of US and in Boston at the "AQ super site". For Europe, the period of study has been chosen from June 20th to July 3rd 2009 with regards to pollution events from 25th to July 2nd observed by AIRPARIF and during MEGAPOLI and shown by MODIS AOD's. **AODs exploitation**: The second part of observations task concern AOD retrievals from satellites: MODIS, CALIPSO and inferring PM2.5 map using statistical model developed by HSPH. MODIS AOD data (public access) are stored by MODIS team at 10km-10 km resolution (provided by a first NASA-MODIS team), twice a day (morning TERRA and afternoon AQUA). The corresponding periods are downloaded for the project objectives. The retrievals used look-up tables for identified composition of aerosols. The 10km-10km AOD resolution is appropriate to study regional scale modelling on D02 (and D01) in this project for US&EU. For urban/local scales, higher resolution is required. A new method recently published by another NASA MODIS team to retrieve AODs, leads to interesting features by producing 1km-1km resolution of AOD, but uncertainties are still under investigation. The fellow cooperation with this second team allows a high resolution AOD mapping over Europe from January to July 2009 from MODIS. NASA AOD at 1km database is available for the project but not for the public use. Days corresponding to the period of investigation and showing high levels of AODs are converted to PM2.5 map concentration using Harvard SPH tools and analyzed versus models results. ***Intermediate conclusion:*** the holistic tool proposed in this project is managed accordingly and is applied for specific cases in US (for hand-on experiences) and EU (return phase) to demonstrate its feasibility, its performance and may be applied to any other multiple scale AQ studies e.g. hot spot pollution events, or (in the future) for much longer model studies of population exposure (trends) such as seasonal to annual trends. This holistic tool may be also used to support abatement strategies, along with policy, of emissions, knowing that all emission sectors are treated separately in the tool; their individual contribution may be discriminated and assessed. Nonetheless, multiple mandatory model evaluations must be performed against available measurements including satellites; this constitutes a common feature for any AQ model applications worldwide and the relationship health to binding cap and exposure better.

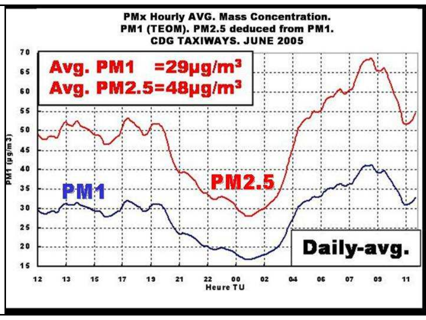
e) Some pertinent scientific results about multi-scale AQ modelling: Measurements obtained during field campaigns at intra-muros (IM) CDG airports for which the fellow was the PI, for PM2.5, PM1, Ozone, CO, NO, NO2, SO2, VOCs and other species were analyzed against monitored PM2.5 by AIRPARIF in Paris IM and along circular highways at the same periods. Results (2005) show that daily averaged PM2.5 in Paris IM is much lower than at airports IM. PM2.5 along circular highways is a bit lower than at airports IM. Ozone concentration in Paris and rural stations are at higher concentration than at airports IM due to higher NOx at airports IM. Airports are generally not very far from mega-cities city limits and should be taken into consideration for air pollution studies. **Weather forecast experiments**: Precise multiscale prediction of AP concentration requires adequate and very good quality of meteorological fields constraining the AQ model, these parameters include: high pressure stagnation duration, air temperature (T), wind characteristics (U), relative humidity (RH), water vapour (Q), quantity of rainfall (R), depth of the boundary layer (DBL) and many other parameters. Model results show that the RH has a net effect on modelled PM2.5 and rainfall acting as losses by wet deposition of its components. WRF results (US, EU) have been analyzed in detail before transformed as input of the AQ model. **Emission database creation :** The discrimination of emission source of species plays different roles on the modelled pollutants. Terpenes from biogenic sources compete with anthropogenic sources (urban) top form organic compounds composing PM2.5. Near higher source emissions such as forests, model shows that biogenic contribution is much higher. In this work, it is demonstrated that emissions spatial resolution are relevant to model and capture small-scales and hot spot AP concentration. Model shows that O3 is sensitive to biogenic reactive VOC and distribution of isoprene produced by forests. Power plants source emissions in mega-cities lead to high concentration of AP and PM2.5 within the cities and well captured by the model but at high resolution domain (D04). **Model vs. observations**: Modelled and observed PM2.5 composition (East of US) is dominated by sulphate, the South East partly by organic compounds (OC) due to biogenic emissions (forests). In Europe, sulphate is the most dominant PM2.5 components; modelled OC (composing PM25) are overestimated due to high biogenic emissions. Regional contribution of biomass burning (BB) emissions to local PM2.5 concentration (modelled) is not very significant. Large-scale transport of aerosol plumes from mega-cities is observed from MODIS (high AOD value) for US and Europe. Regional polluted air of O3 and PM2.5 formed under stagnant atmosphere, observed at AQ stations and fingerprinted by inferred PM2.5 from AOD's match well with model results for US. Ground observations at Boston HSPH supersite are analyzed against numerical results showing that pollutant plumes (PM2.5 incl.) from power plants in the city are well captured by the model at high resolution only (D04 and D03). In Europe, modelled PM2.5 map distribution is consistent with MODIS AOD, maximum of PM2.5 are calculated over mega-cities in Europe: the Netherlands Coastline, Germany, Belgium, North Coast of UK, North of Italy, Center of France. However, PM2.5 daily average is overestimated by the model as well as O3 in Paris center when compared to AIRPARIF measurements likely related to anthropogenic emissions. Throughout Europe, modelled PM2.5 is overestimated also in regions with maximum of terpene emissions (estimated by MEGAN), sources of OC. NH3 emissions (agriculture, farms, and urban) effects on modelled PM2.5 leading to ammonium sulphate and nitrate, are significant in Europe. Generally (US and EU), the model shows some deficiencies on plume dispersion and mixing (such as from chimneys) and on the fate of emitted species (e.g. NO/NO2 equilibrium), oxidation is likely too slow. It is demonstrated that urban scale air pollution prediction for all AP matches better with observations with higher resolution (D04-D03) and the interaction of scales are also pertinent e.g. effects of regional (D02) transport to local scales (D03 and D04). In early morning, due to high volume of emission by heavier ground traffic and other sources at the time where the boundary layer depth is thin, accumulation of PM2.5 is predicted at this time unlike ozone which is photo chemically formed at midday. In the afternoon, WRF predicts a deeper boundary layer so that AP are mixed with altitude and less accumulated. Modelled O_3 is maximum at 14-15hLoc during hot spots whereas other AP e.g.: SO2, NO2, PM2.5 and its composition fits more with emission diurnal variations. **The holistic tool**, particularly the AQ model constrained by the weather forecast meteorology, shows a performing capability to predict aerosols characteristics relevant for AOD retrievals from satellites including: number size distribution; chemical composition: inorganic, organic, soot; effects of RH, effect of particle mixing state (internally or externally mixed). Consequently, the tool is able to provide more realistic, complete and precise look-up tables of aerosol characteristics for AOD retrievals. Inferring PM2.5 maps from AOD from satellites is very dependent on cloud cover, over which, AOD maps are not produced. In this case, inferred PM2.5 show gaps where clouds are present. This is a serious limitation of the use of AOD to infer PM2.5 for regular monitoring. But a combination of ground data, satellite, model results (as in this tool) are very promising for PM2.5 characterisation over broad regions and mega cities .These results have been presented at peer-reviewed high-level Conferences and some workshops. Some final peer-reviewed publications to scientific journals are under preparation with deeper analyses of results and will be reported as indicated by the EC on participants' portal. Some non-confidential results will be stored on the project web site for public access.

General Conclusion: Regardless of significant computer system issues encountered by the fellow in US and EU; the non-existence of adequate multiscale emission databases in Europe, the holistic tool is elaborated, tested, applied and evaluated for US and EU continents from regional to local-urban scales down to mega-cities. A large number of cooperation is initiated in this work, will continue and be strengthened for: satellite data exploration, emission processing, modelling (weather and air quality), ground observation analyses, lecturing (Harvard Univ.); bringing pertinent added values to characterise PM2.5 and its hazardous chemical composition along with O3 in Europe. This project proposes a new strategy of air pollution investigations in Europe based on multiscale approach relevant for the interaction of emission origins and AP lifetime. **Socio-economic impact**: as the contribution of each sector of emissions on AP concentration can be discriminated by the tool, assessment studies may be applied to support policy requirements, mitigation options and abatement strategy. Nevertheless, the tool evaluation must be appropriately multiplied. In addition, its ability to map PM2.5 (time and space) and its hazardous chemical composition at different scales constitute a relevant pre-requisite for population exposure and epidemiological studies. **Targets:** policy, population, emission sectors, scientific groups.

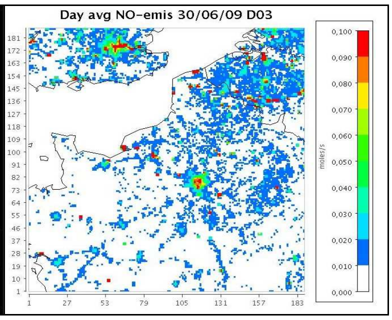
APPENDIX



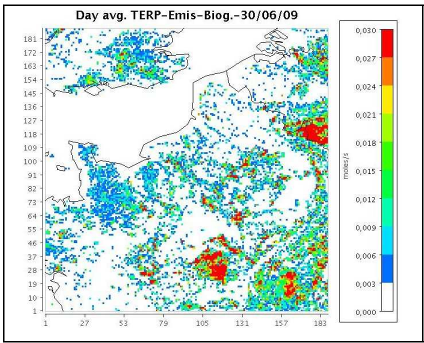
Obs. Avg. PM2.5. Paris mega-city.
Intra-muros- Highways (same period)



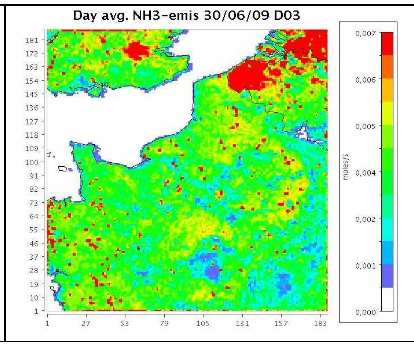
Obs. Avg. PM2.5 Paris CDG airports
Intra-muros (same period)



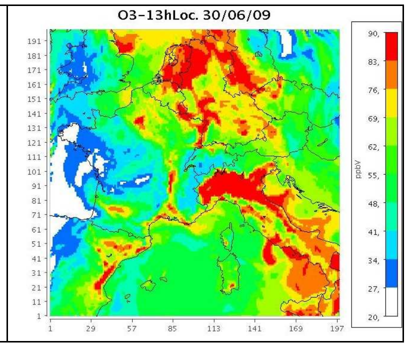
Anthropogenic NO emiss. Dom3
Calculated w/t SMOKE-EU



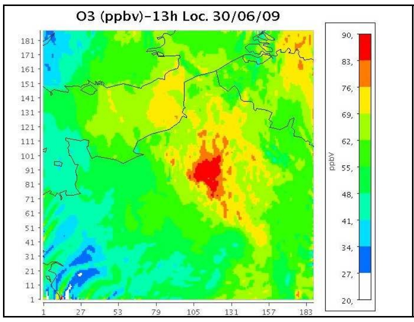
Biogenic Terpene emissions
Source of Organic Compounds.
for PM2.5 composition. EU

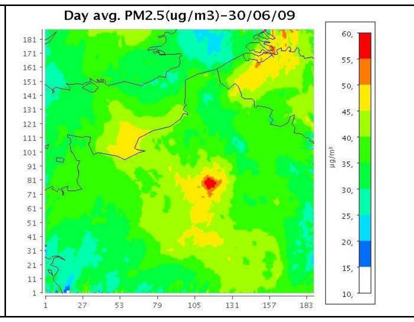


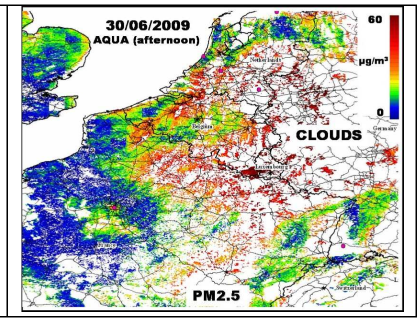
NH_3 emissions all inclusive:
Agriculture, farms, power plants.
for PM2.5 composition. EU



O_3 map 13HLoc (modelled)
Regional scale domain 2.
EU



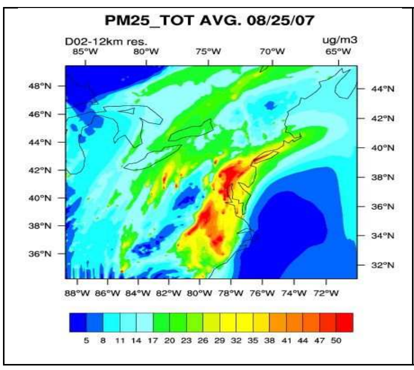




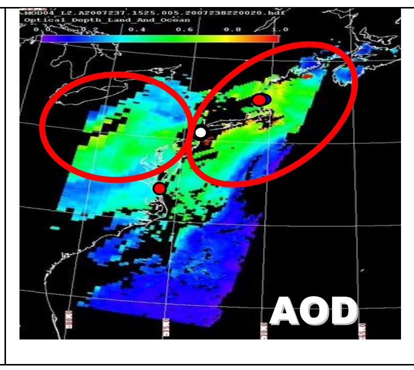
O_3 map 13hLoc (modelled)
Local-urban domain 3. EU

PM2.5 Daily Avg. (modelled)
Local-urban domain 3. EU

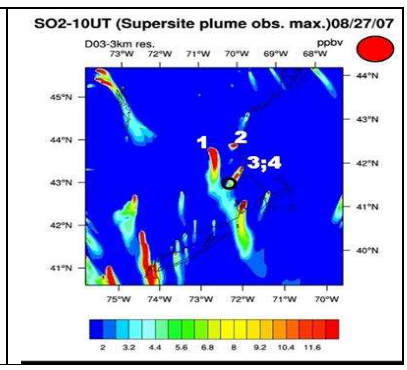
Inferred PM2.5 from MODIS AOD
1km res. (white=no data)
(no data over ocean)
Statist. model w/t ground obs.



PM2.5 map day average (US-East)
Modelled. Regional domain 2. US



MODIS AOD (08/25/07). US
shows same features than PM2.5
10km by 10km (regional)



Boston (US-dom4) SO_2 plumes(modelled)
Power plant sources. High res. 1km
Consistent with HSPH observations.
Circled: AQ station. N°: Point sources