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Inverse modeling of particulate matter sources in SE Central Europe

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INTRODUCTION

Atmospheric trace substances include particulate matter (PM) and gases. PM and gases originates from both natural and anthropogenic emission sources. However, while anthropogenic emission sources are driven by human and industrial activities, natural emissions are very dependent on seasonal changes and multi-year climate changes. The most common anthropogenic emissions are PM and various gases (CO2, CO, NH3, SO2, NOX, …). In addition to Greenhouse Gases (GHG), PM plays an important role for the radiative budget of the Earth. PM both scatters/reflects and absorbs solar radiation. Additionally, PM is a major component of air pollution and has great effect on human health. PM air pollution is mainly a regional issue as pollutants released in one country can be transported in the atmosphere affecting air quality in the nearby countries. When implementing air quality management strategies, detailed information about local and regional anthropogenic and natural emission sources are desired. Some locations are more affected by local emissions, however other locations are more affected by long-range transport because of unfavorable meteorological conditions. Particulate matter and greenhouse gases are commonly co-generated by the combustion of fossil fuels, and therefore additional knowledge and more accurate emission inventories of PM will provide insight into GHG emission inventories. To achieve necessary air pollution reductions and in preparation for future agreements on climate change both air quality improvements and GHG reductions are needed.

AIM

During past years the density of the ground-based network of measuring stations to systematically measure concentrations of pollutants in ambient air was increased. Air pollution data measured at site specific stations can be used to extract source emission properties. This optimization technique is called inverse modeling (Seibert 2000, 2001). The goal is to adjust emission sources to minimize the differences between model output and real measurements. In the presented research work the transport of the PM10 emissions will be simulated with the Lagrangian particle dispersion model FLEXPART (Stohl et al., 1998, 2005). The main aim of this work is to gain knowledge of inverse modeling methods using the Lagrangian model FLEXPART with emphasis on data pre-processing, setting model parameters and to set up inversion.

EXPLORATORY DATA ANALYSIS

Observations

The European air quality database (http://www.eea.europa.eu/) consists of a multi-annual time series of air quality measurement data and covers geographically all EU Member States, the EEA member countries and some EEA collaborating countries. Matlab and Shell scripts were developed to extract data according to time period, completeness and type of station. We chose daily data because there were many more data stations available. The selected period for analysis and for inverse modeling input data was set to the period from beginning of year 2010 to the end of year 2012. In Figure 1 left, all stations (1396) are plotted, which have completeness above 90% and in Figure 1 right, stations (158) are shown, which are categorized as background rural and have completeness above 90%.

As reported by Kaiser *et al*, 2007 and Spangl *et al*, 2006 Austria is affected by long range transport from nine regions: 1) North Italy 2) Slovenia, Croatia, Bosnia, 3) Serbia, 4) Romania, 5) Hungary, Slovakia, 6) East Poland, West Ukraine, East Slovakia, Hungary, 7) South Poland, 8) Poland, Czech Republic, East Germany, 9) England, Benelux, North and East France, Northwest, West, Central and South Germany, North Switzerland. The possible main sources of PM10 are thus air masses with a long residence times over the central, northwest and northeast Europe and air masses from the west and southwest, which are favored by a foehn effect crossing the Po Basin.

Accordingly, the Obershützen station was as the reference station and cross-correlations with other stations were calculated. Figure 1, right, shows the correlations between Obershützen station and all other background stations. It can be seen that the correlation drop with distance. The maximum correlation was 0.86 (HU0040A, Sarrod, lon=16.838896, lat=47.671947) and the minimum correlation was -0.24 (PT04006, Terena, lon=-7.397514,lat=38.614998). It follows that more distant stations are less correlated.

In addition to correlation analysis, clustering analysis was performed. The clustering was conducted by using k-means, where three different similarity measures were tested (Theodoridis and Koutroumbas, 2008): a squared Euclidean distance, a city-block distance and a cross-correlation. The k-means clustering with the city-block distance was chosen as the default clustering. For the selection of a proper number of clusters in the default k-means clustering two tests were carried out, one by observing the within-cluster sum of squares values against the number of clusters and the other by performing silhouette-value analysis (Kaufman and Rousseeuw, 1990). Figure 2 shows five clusters of rural background stations. Typical clusters profile are presented in Figure 3 (top row), meanwhile Figure 3 (bottom row) shows auto-correlation of cluster profiles. This analysis shows that clusters 1, 2, 3 have seasonal pattern with high concentrations during winter and low concentrations during summer time. Cluster 3 has a very pronounced seasonal variation; the stations of these clusters are mainly located in northern Italy (Po Basin). Clusters 4 and 5 show poor seasonal variation and these stations are mainly located in the western Mediterranean, in Spain and in Portugal.

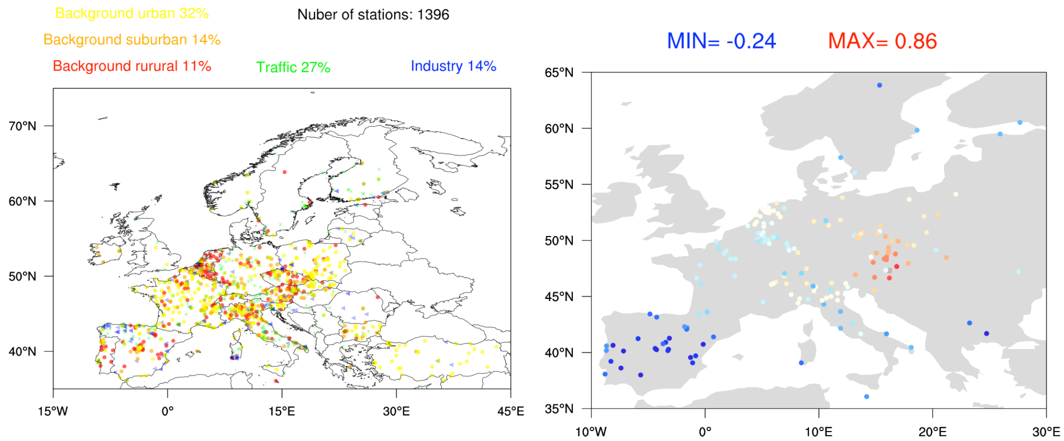

Fig1. Ground base stations over Europe. Left, all stations that have completeness of data above 90%, right; rural background stations.

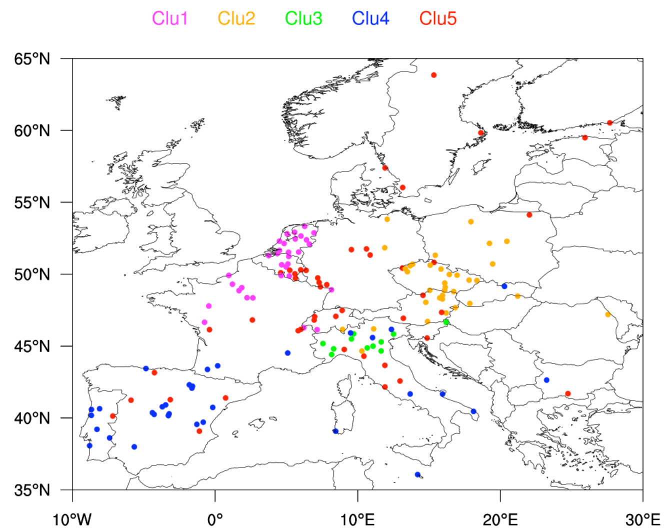

Fig.2 Clusters of rural background stations.

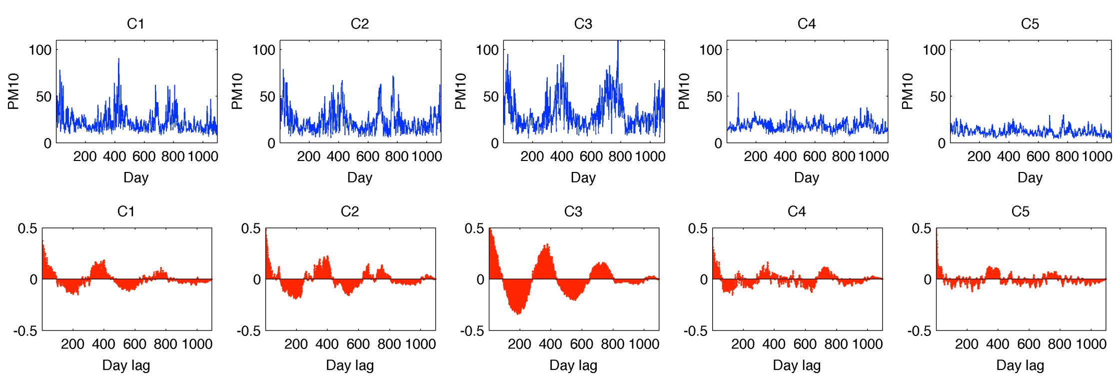

Fig.3 Typical profiles of clusters and auto-correlation of clusters.

Prior emission fluxes

Particulate matter originates from both natural and anthropogenic emission sources. However, while anthropogenic emission sources are driven by human and industrial activities, natural emissions are very dependent on seasonal changes and multi-year climate changes. Weather has a strong influence on the natural sources of emissions, for example, windblown crustal and sea-spray particles. The prior fluxes were composed from the EDGAR (http://edgar.jrc.ec.europa.eu/) and MACC-II databases (Granier et al., 2012; Kuenen et al., 2013;). In air quality modeling these emissions are divided over the year using monthly, weekly and hourly time profiles to reflect variations in activities that cause emissions.

The integrated PM10 emissions over all sectors of the EDGAR data emission inventory database are shown in Figure 4. The EDGAR database involves various emissions categories:

- agricultural soils,
- agricultural waste burning,
- aviation,
- energy manufacturing transformation
- fossil fuel fires
- fugitive from solid
- international navigation
- industrial processes and product use
- livestock
- non-road transportation
- production of oil and gas
- residential and other
- road transportation
- waste

In different countries different life styles, habits, holidays and meteorological conditions are present and consequently prior emissions fluxes are not constant. Figure 5 shows typical daily, weekly and seasonal temporal profiles. Hourly fluxes are highest during morning and evening rush hour, weekly temporal profiles shows that lowest fluxes are during weekend when there is lower traffic density and less industrial activity. Seasonal profiles show the lowest concentration during hot periods and are higher during spring and autumn. The spring and autumn periods include more intense agricultural activities, meanwhile during winter periods higher emissions are due to a domestic heating.

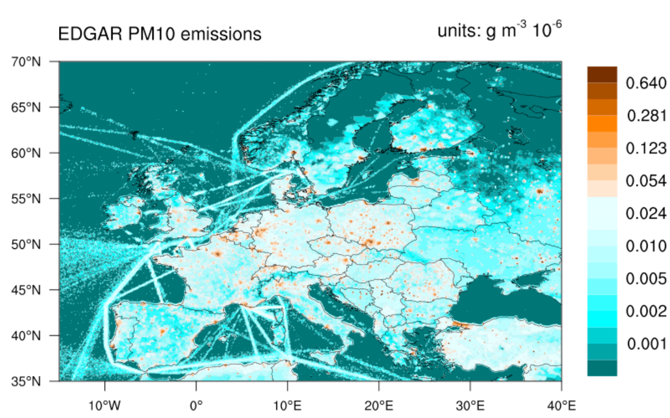

Fig.4 Edgar emissions fluxes integrated over all sectors.

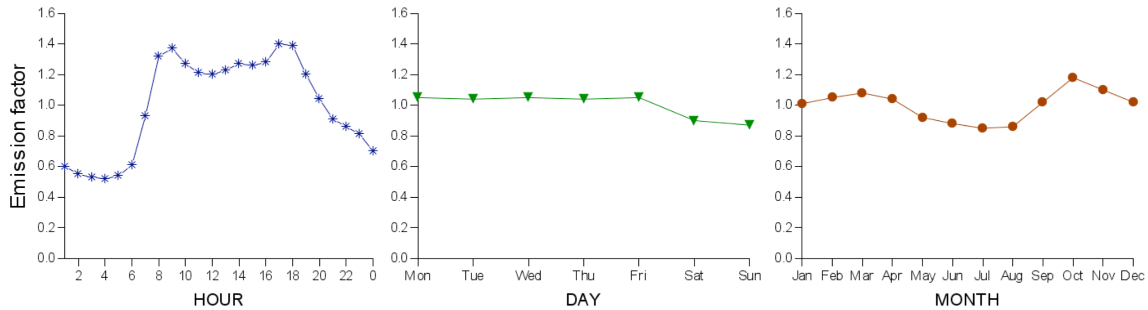

Fig.5 Time profiles for PM2.5 particles.

3D concentration fields

When the inversion method is performed, 3D fields are need for calculation of the initial mixing ratios. The MACC (https://www.gmes-atmosphere.eu/) data are produced at 4 levels: surface, 500, 1000, 3000 meters. The MACC ensemble is currently based on the median method of seven model forecasts: CHIMEREby INERIS (France), EMEP by MET Norway (Norway), EURAD-IM by University of Cologne (Germany), LOTOS-EUROS by KNMI (Netherlands), MATCH by SMHI (Sweden), MOCAGE by Météo-France (France) and SILAM at FMI (Finland). The seven regional air quality models provide daily forecasts of the concentration of the main atmospheric pollutants in the lowest atmospheric layers for the following 4 days. Their horizontal scale is around 10 to 20 km, thus representing large scale phenomena and the background air pollution. In Figure 6 concentrations of MACC surface level for four seasonal periods are presented: December-January-February (DJF), March-April-May (MAM), June-July-August (JJA) and September-October-November (SON).

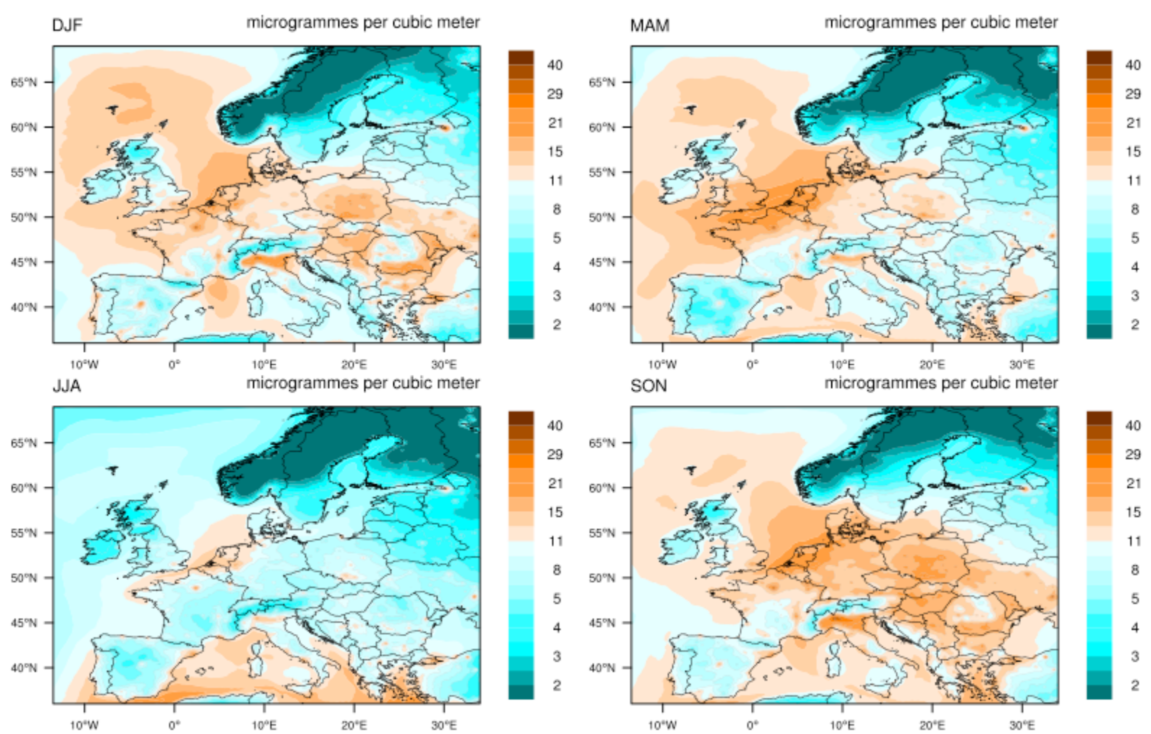

Fig.6 Seasonal averages of PM10 over Europe at surface level.

ECMWF meteorological data and FLEXPART set up

To generate a source-receptor relationship the Lagrangian particle-dispersion model FLEXPART was used (Emanuel and Zivković-Rothman, 1999; Stohl et al., 1998; Stohl et al., 2005; Stohl and Thomson, 1999). The test run was driven by ECMWF data with the temporal resolution of a 3-hour forecast at 00:00, 03:00, 06:00, 09:00, 12:00, 15:00, 18:00, and 21:00 UTC; the horizontal resolution was $0.25^o \times 0.25^o$ and the vertical resolution was 91 layers. The ECMWF data were retrieved using EMWFDATA V6.0 software. The FLEXPART was run in a backward mode. There was a horizontal resolution of $0.1^o \times 0.1^o$ (nested) and of $0.5^o \times 0.5^o$ in one vertical level at 100, 500, 1000 and 3000 meters. In backward mode 500000 particles were released from the release point, followed backwards in time, calculating the response function that is related to the particle's residence time (Seibert, 2000, 2001). FLEXPART was set up to generate 3-hourly trace releases from the measuring location and the output was produced on the regional out-grid domain (-10E/35E/35N/60N). The particles were released as a FLEXPART pre-defined SO_2 aerosol tracer and a maximum particle age class of 7 days was allowed.

At this stage of research work the FLEXPART was run for limited number of stations and time period. To estimate the time needed and how many stations can be used for inversion, we ran FLEXPART for four stations for period from 30. 4. 2012 to 1. 7. 2012. Test runs showed that when using 500000 particles released each day then duration of the run was approximately 2 hours. Figure 7 shows a footprint (level at 100m) of Obershützen (Austria), Dolny Studenky (Czech), Iskrba (Slovenia) and Toplniky (Slovakia) station.

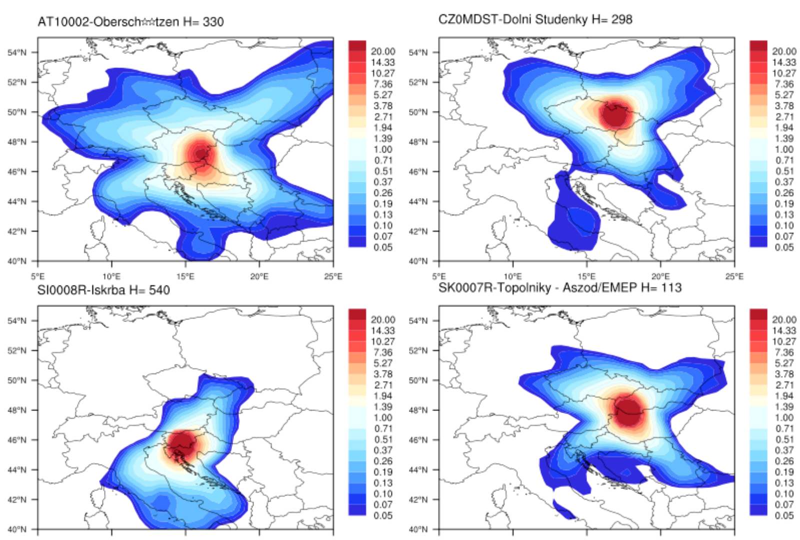

Fig.7 Footprint of Obershützen (Austria), Dolny Studenky (Czech), Iskrba (Slovenia) and Topolniky (Slovakia) station in southeast central Europe for period from 30. 4. 2012 to 1. 7. 2012.

CONCLUSION

The main aim of this Exchange grant was to gain knowledge of inverse modeling methods using the Lagrangian model FLEXPART. During the exchange visit special attention was given to data pre-processing and exploration analysis of available PM10 data. It can be conclude that data of AirBase was explored in detail and that data were set up to run FLEXPART model. The extended plan is to calculate source receptor relationships for at least 10 ground-based release points for a period of two years. The estimation is that calculation of source-receptor relations ship will take approximately 2 months. After obtaining source-receptor relationship data we have planned to use FLEXINVERT (Thompson and Stohl, 2014), which is a Bayesian inversion framework for optimizing surface-to-atmosphere fluxes of atmospheric species as long as the chemistry can be described as a linear process.

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