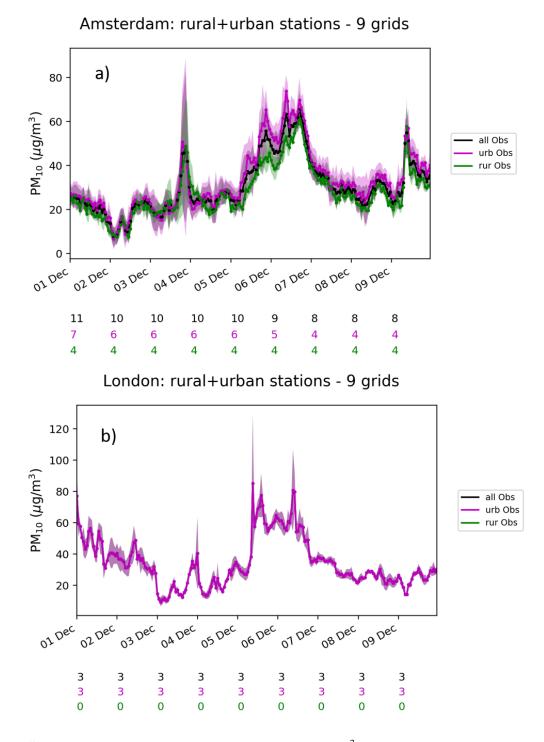
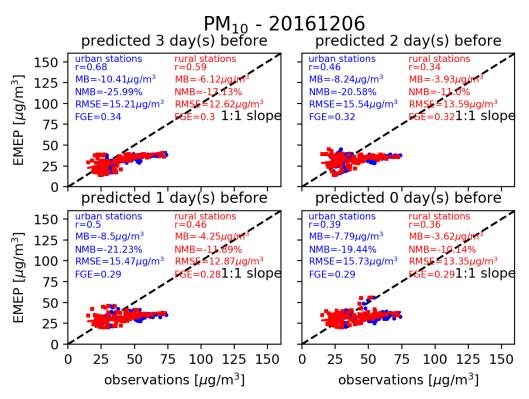


Figure. S1 Boxes defining each city edge, based on the 1 grid, 9 grids and the GADM definitions.



**Figure. S2** Hourly averaged  $PM_{10}$  concentrations (in  $\mu g/m^3$ ) measured by the AirBase stations in Amsterdam (a) and London (b) from Dec  $01^{st}$  to  $09^{th}$  2016. The cities are defined by an area using 9 grid cells. The mean of all the stations is plotted in black, the urban stations are plotted in magenta and the rural stations in green. The colored shade error corresponds to the standard deviation. The colored numbers below the time-series correspond to the number of stations per day.



**Figure. S3** Scatterplot between the hourly  $PM_{10}$  concentrations over all the studied cities using the 9 grids definition, predicted by the EMEP model on December 06<sup>th</sup> 2016 and the observations of the urban sites (blue dot) and rural sites (red square). For this case, only the five cities having urban and rural stations are used. The observations are collocated in time to the EMEP predictions and then averaged within the city edge to match the studied grid. The four panels correspond to the different predictions from 3 days before the December 06<sup>th</sup> to the actual day, i.e. December 06<sup>th</sup>. The correlation coefficient (r), the mean bias (MB), the normalized mean bias (NMB), the root-mean-square error (RMSE) and the fractional gross error (FGE) are provided on each panel. The blue and red lines represent the linear fits.

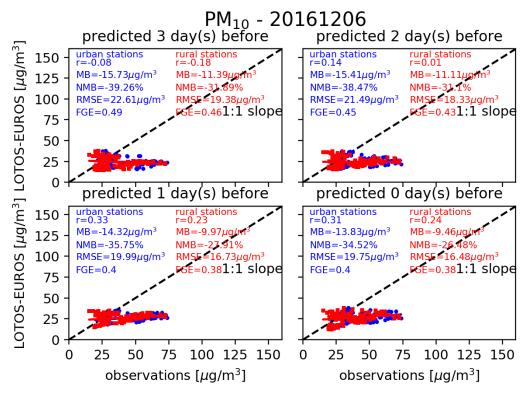
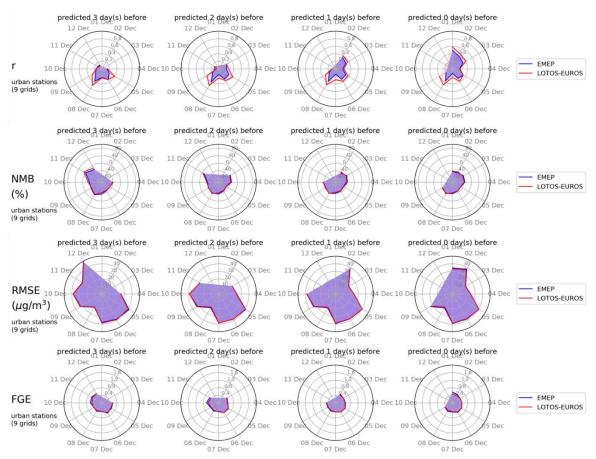


Figure. S4 As Fig. S3 for LOTOS-EUROS



**Figure. S5** Spider plot presenting the correlation coefficient, the normalized mean bias, the root-mean-square error and the fractional gross error for all the predicted days (from 01 to 12 December – with starting dates from 01 to 09 December) over the cities defined by 9 grids. In maximum, there are 19 cities represented. The blue shade and line represent the EMEP performances and the red blue shade and line represent the LOTOS-EUROS performances. The four spider plots on each line present the results depending on the starting date of the forecast. The negative correlations are not shown.

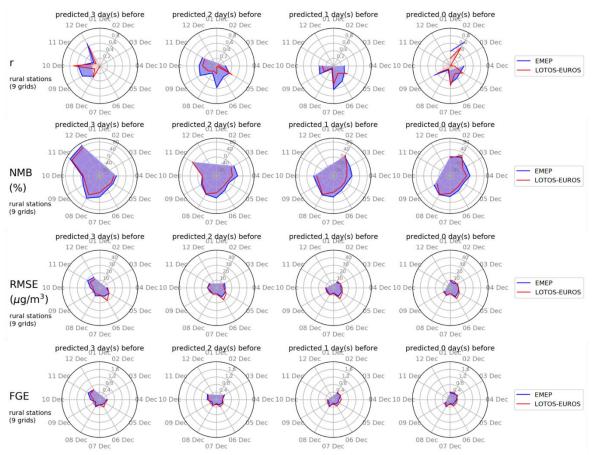
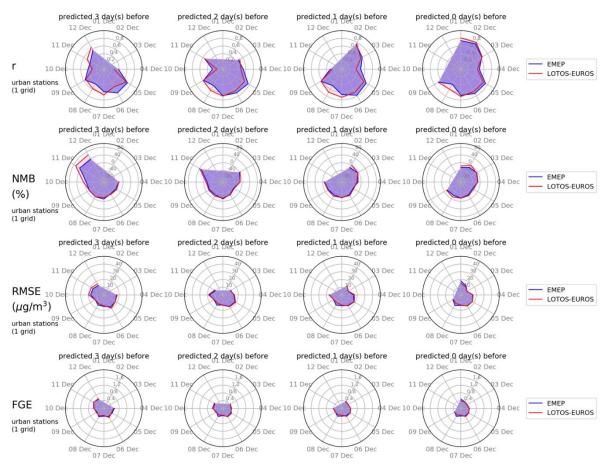
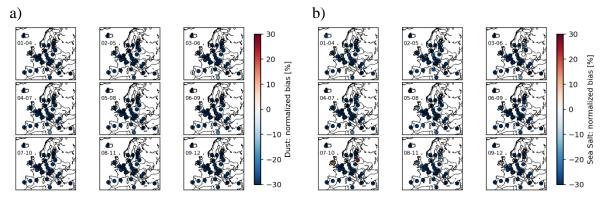


Figure. S6 As Fig. S5 for the rural stations. In maximum, there are 5 cities represented.

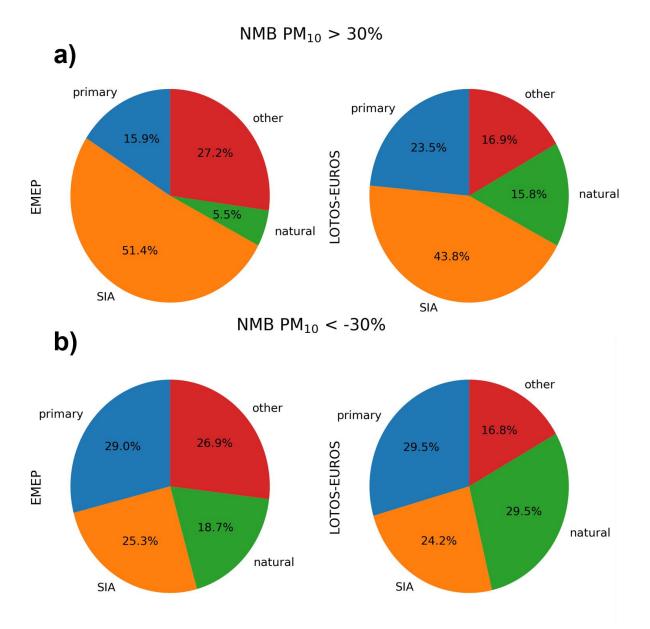


**Figure. S7** Spider plot presenting the correlation coefficient, the normalized mean bias, the root-mean-square error and the fractional gross error for all the predicted days (from 01 to 12 December – with starting dates from 01 to 09 December) over the cities defined by 1 model grid. In maximum, there are 16 cities represented. The blue shade and line represent the EMEP performances and the red blue shade and line represent the LOTOS-EUROS performances. The four spider plots on each line present the results depending on the starting date of the forecast.

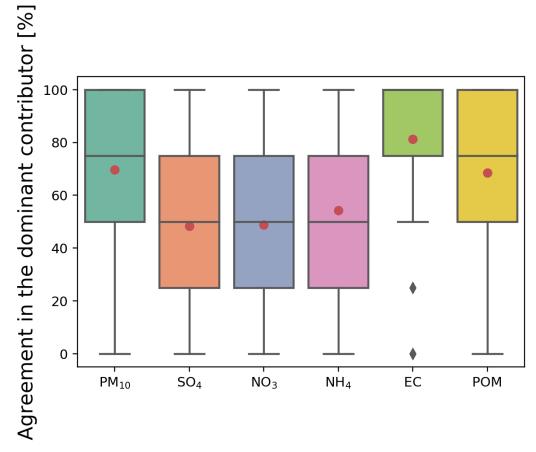


**Figure. S8** Normalized mean bias between the EMEP model and LOTOS-EUROS for dust (a) and sea salt (b) over the 34 European cities using the 9 grids definition for each 4day-forecast (01-04 Dec 2016, 02-05 Dec 2016, 03-06 Dec 2016, 04-07 Dec 2016, 05-08 Dec 2016, 06-09 Dec 2016, 07-10 Dec 2016, 08-11 Dec 2016, 09-12 Dec 2016). The NMB is calculated as:  $\Sigma(EMEP-LOTOSEUROS) \times 100\%$ .

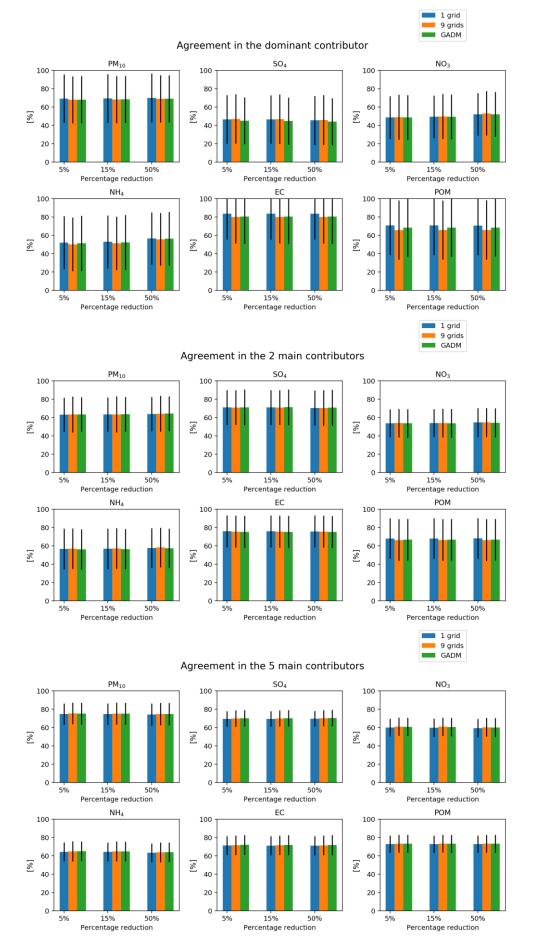
 $\sum LOTOSEUROS$ 



**Figure. S9** Mean distribution in percent of the  $PM_{10}$  components based on the NMB of the  $PM_{10}$  predicted over the 34 European cities using the 9 grids definition for each 4day-forecast. a: Only the distribution of the components for the cities having a NMB larger than 30% is shown. b: Only the distribution for the cities having a NMB lower than -30% is shown. "Other" is calculated as the difference between the  $PM_{10}$  concentrations and the sum the three other components (primary = POM+EC, SIA and natural).



**Figure. S10** Agreement in the determination of the daily dominant country contributor for  $PM_{10}$ ,  $SO_4$ ,  $NO_3$ ,  $NH_4$ , EC and POM in percent, determined over all the studied cities using the 9 grids definition and for all forecasted days. The line that divides the box into two parts represents the median of the data. The end of the box shows the upper and lower quartiles. The extreme lines show the highest and lowest value excluding outliers which are represented by grey diamonds. The red dots correspond to the mean of each data set.



**Figure. S11** Mean agreement between both SR calculation methodologies in the determination of the dominant country contributor, the two main contributors and the five main contributors for  $PM_{10}$ , SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>, EC and POM in percent, determined over all the studied cities and for all 4day-forecasts. The results for the 3 city definitions (1 grid, 9 grids, GADM) and for the percentage of reduction used in the perturbation EMEP runs (5%, 15%, 50%) are shown. The black lines correspond to the standard deviation.