Suggestions for revision or reasons for rejection (visible to the public if the article is accepted and published)

Authors introduced relevant updates and improvement with respect to both methodological description and presentation of the obtained results.

However the comparison of modelled concentrations (A*x) against observations point out a clear and systematic underestimation.

Authors state that such underestimation indicates that "most of observed concentrations are from local sources, hence they cannot be reproduced by the A*x model.

In my opinion this explanation is rather weak and questionable.

I would ask the authors to review the conclusions and partially the discussion section in order to:

- Better explain and justify the underestimation that affect modelled results

- Discuss which could be, anyhow, the usefulness of such model, although it shows a clear underestimation. This latter answer could take advantage of the arguments of the first answer.

The authors are really thankful to the reviewer for the excellent comments and recommendations.

*An error was found in the estimation of the OMI-HTAP emission map 1°*1° average, which is now corrected.*

Our answers follow in italics:

1. Better explain and justify the underestimation that affect modelled results

We will update the Tikhonov regularization equation in the manuscript, including the a priori information on emission fluxes from each grid cell (x_0). The equation follows:

$$min\{||Ax-b||^2 + \lambda^2||L(x-x_0)||^2\}$$

When no a priori information is available, we make the assumption in the Tikhonov regularization equation that x_0 is 0. In other words, our a priori information is that the emission fluxes from all grid cells are 0. We seek in our case a smooth solution, requesting that emission fluxes of neighboring cells have 0 differences. Smaller differences are achieved when absolute emission fluxes values are small (closer to 0). This imposes solutions with as small as possible emission fluxes, leading to the underestimation of measured values. The underestimation is relevant to how much we have to depart from the perfect fit (the fidelity term) of the measured data in the regularization procedure, which is achieved when the regularization parameter λ is equal to 0. As we mention in section 2.1 (PM sampling stations and filter analysis), due to the high number of cities involved in the study, it was not possible to fully harmonize the methods used. In section 2.3 (Tikhonov regularization) we mention that due to positive and negative artifacts in the filter sampling, uncertainties in the filter analysis and PMF, an overall uncertainty approximating 30% is expected. In order to regularize such an uncertainty level, a large regularization parameter λ is required. Thus, the regularization term is very important, leading to an underestimation of the resulting emission fluxes. We have to keep in mind that if λ is close to 0, we perfectly reconstruct the measured concentrations, which include a

large error due to the reasons mentioned earlier. As the inverse process is not linear, such an approach would result in very large errors in the estimation of emission fluxes in each grid cell.

We will update in the manuscript in section 2.3 (Tikhonov regularization) the adjusted equation and replace lines 162-168 with the following:

"When no a priori information is available, the assumption in the Tikhonov regularization equation is that x_0 is 0. We seek in our case a smooth solution, requesting that emission fluxes of neighboring cells have 0 differences. Smaller differences are achieved when absolute emission fluxes values are small (closer to 0). This imposes solutions with as small as possible emission fluxes, leading to the underestimation of measured values. The underestimation is relevant to how much we have to depart from the perfect fit (the fidelity term) of the measured data in the regularization procedure, which is achieved when the regularization parameter λ is equal to 0. As we mentioned in the previous paragraph, an overall uncertainty approximating 30% is expected. In order to regularize such an uncertainty level a large regularization parameter λ is required, thus leading to a significant underestimation of the model results. We have to keep in mind that if λ is close to 0, we perfectly reconstruct the measured concentrations, which include a large error due to the reasons mentioned earlier. As the inverse process is not linear, such an approach would result in very large errors in the estimation of emission fluxes in each grid cell."

2. Discuss which could be, anyhow, the usefulness of such model, although it shows a clear underestimation. This latter answer could take advantage of the arguments of the first answer.

The new method was developed in order to contribute to the air quality management process.

The usefulness of the proposed two-step method lies in the following points:

1. *Improved identification of source areas for the long range transported aerosol in comparison to PSCF analysis.*
2. *Classification of relative importance of emission fluxes from geographic grid cells. This classification could be compared to existing emission inventories, resulting in possible improvements in the emissions calculation algorithms.*
3. *Estimation of the magnitude of emission fluxes from each grid cell. For Secondary Sulfate, around 60% of the measured concentrations magnitude could be reconstructed based on the deducted emission fluxes, while for Dust, approximately 45% could be reconstructed. This indicates that in this case, the new method significantly underestimates emission fluxes and measured concentrations. We have to keep in mind though that if data with much lower uncertainty are used, the underestimation would be significantly lower. Also, additional a priori information could lead to better performance of the method.*
4. *Since we identify the pollutant source area, its relative magnitude and acquire an estimate of the measured concentrations reconstruction, we can implement targeted mitigation measures. This approach can be used for any pollutant that can be simulated in FLEXPART or any similar model, without the need of an emission inventory.*
5. *Ideally, we would like to use the new method in combination to chemical transport models, so as to improve mitigation measures estimation. We should keep in mind that the emission fluxes deducted by the new method are averages over a period of 3 years. Emission fluxes have seasonal, monthly, weekday and daily variations in each grid cell that could not be*

identified. Therefore, the emission fluxes result derived by the new method can only approximate roughly the concentrations measured at the cities participating in the study.

We will add in the manuscript as Appendix A5 the figures for Secondary Sulfate and Dust, where slopes between modeled and measured concentration without intercept are depicted. We will mention these figures in section 4 (Summary and Conclusions).

We will also add in section 4 (Summary and Conclusions, lines 393-409 in the revised manuscript) the following:

"The authors developed the new method in order to contribute to the air quality management process.

With this new method, an improved identification of source areas for the long range transported aerosol in comparison to simple PSCF analysis is achieved. Also, the relative importance of emission fluxes from each geographic grid cell is classified. This classification could be compared to existing emission inventories, resulting in possible improvements in the emissions calculation algorithms.

The new method also provides an estimate of the magnitude of emission fluxes from each grid cell. For Secondary Sulfate, around 60% of the measured concentrations magnitude could be reconstructed based on the deducted emission fluxes, while for Dust, approximately 45% could be reconstructed. This indicates that in this case, the new method significantly underestimates emission fluxes and measured concentrations. We have to keep in mind though that if data with much lower uncertainty are used, the underestimation would be significantly lower. Also, additional a priori information could lead to better performance of the method.

Since we identify the pollutant source area, its relative magnitude and an estimate of the measured concentrations reconstruction, we can implement targeted mitigation measures. This approach can be used for any pollutant that can be simulated in FLEXPART or any similar model, without the need of an emission inventory.

Ideally, we would like to implement the new method in combination to chemical transport models, so as to improve mitigation measures estimation.

We should keep in mind that the emission fluxes deducted by the new method are averages over a period of 3 years. Emission fluxes have seasonal, monthly, weekday and daily variations in each grid cell that could not be identified. Therefore, the emission fluxes result derived by the new method can only approximate roughly the concentrations measured at the cities participating in the study."