Project: **1260**

Project title: **Megacity Aerosol Composition by Satellite: A tool to study anthropogenic Emissions, Climate change and human Health**

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Report period: **2023-11-01 to 2024-10-31**

The allocated resources were used for the analysis of the two EMeRGe aircraft campaigns conducted in Europe from July 11 to 28, 2017, based in Munich, Germany, and in East Asia from March 8 to April 9, 2018, based in Taipei, Taiwan.
As explained in the previous year's report, an article was about to be submitted to ACP. Finally, two articles have been submitted that are currently under review and we hope to finalize the review process in early 2025.

1) The first article **egusphere-2024-516** is entitled: ***Air quality model assessment in city plumes of Europe and East Asia.***
https://egusphere.copernicus.org/preprints/2024/egusphere-2024-516/

2) The second article **egusphere-2024-521** is entitled: ***Proportional relationships between carbonaceous aerosols and trace gases in city plumes of Europe and East Asia.***
https://egusphere.copernicus.org/preprints/2024/egusphere-2024-521/

Therefore, we did not have to add new sensitivity experiments but it is important for us to keep the possibility of doing so as needed to respond to the reviewer during the review process, and also to access our data until the end of the review process. In addition, the great support of DKRZ is acknowledged in both articles.

Article 1: *Air quality model assessment in city plumes of Europe and East Asia*

Abstract

An air quality model ensemble is used to represent the current state-of-the-art in atmospheric modeling, composed of two global forecasts and two regional simulations. The model ensemble assessment focuses on both carbonaceous aerosols, *i.e.* black carbon (BC) and organic aerosol (OA), and five trace gases during two aircraft campaigns of the EMeRGe (Effect of Megacities on the Transport and Transformation of Pollutants on the Regional to Global Scales) project. These campaigns, designed with similar flight plans for Europe and Asia, along with identical instrumentation, provide a unique opportunity to evaluate air quality models with a specific focus on city plumes.

The observed concentration ranges for all pollutants are reproduced by the ensemble in the various environments sampled during the EMeRGe campaigns. The evaluation of the air quality model ensemble reveals differences between the two campaigns, with carbon monoxide (CO) better reproduced in East Asia, while other studied pollutants exhibit a better agreement in Europe. These differences may be associated to the modeling of biomass burning pollution during the EMeRGe Asian campaign. However, the modeled CO generally demonstrates good agreement with observations with a correlation coefficient (R) of ≈ 0.8. For formaldehyde (HCHO), nitrogen dioxide (NO_2), ozone (O_3) and BC the agreement is moderate (with R ranging from 0.5 to 0.7), while for OA and SO_2 the agreement is weak (with R ranging from 0.2 to 0.3).

The modeled wind speed shows very good agreement (R ≈ 0.9). This supports the use of modeled pollutant transport to identify flight legs associated with pollution originating from major population centers targeted among different flight plans. City plumes are identified using a methodology based on numerical tracer experiments, where tracers are emitted from city centers. This approach robustly localizes the different city plumes in both time and space, even after traveling several hundred kilometers. Focusing on city plumes, the fractions of high concentration are overestimated for BC, OA, HCHO, and SO_2, which degrades the performance of the ensemble.

This assessment of air quality models with collocated airborne measurements provides a clear insight into the existing limitations in modeling the composition of carbonaceous aerosols and trace gases, especially in city plumes.

Article 2: *Proportional relationships between carbonaceous aerosols and trace gases in city plumes of Europe and East Asia*

Abstract

The concentration of carbonaceous aerosols, black carbon (BC) and organic aerosol (OA), in the atmosphere is related to co-emitted or co-produced trace gases. In this study, we investigate the most relevant proportional relationships between both BC and OA with the following trace gases: carbon monoxide (CO), formaldehyde (HCHO), nitrogen dioxide (NO_2), ozone (O_3), and sulfur dioxide (SO_2). One motivation for selecting these trace gases is that they can be observed using remote sensing measurements from satellite instrumentation, and could therefore be used to predict spatial changes in the amounts of BC and OA.

Airborne measurements are optimal for the analysis of both the composition of aerosols and trace gases in different environments ranging from unpolluted oceanic air masses to those in heavily polluted city plumes. The two aircraft campaigns of the EMeRGe (Effect of Megacities on the Transport and Transformation of Pollutants on the Regional to Global Scales) project have created a unique database, with flight plans dedicated to studying city plumes in two regions, Europe (2017) and East Asia (2018), along with identical instrumental payload.

Using linear regression analysis, three relevant relationships between carbonaceous aerosol and trace gases are identified:

- The BC/OA ratio observed in the Asian campaign is three times higher (≈ 0.3) than in the European campaign (≈ 0.1), whereas the Pearson correlation coefficient (R) between BC and OA is much higher in Europe (R ≈ 0.8) than in Asia (R ≈ 0.6).

- The CO/BC ratio is also observed higher in the Asian campaign (≈ 240) than in the European campaign (≈ 170), whereas the R-value between CO and BC is similar for both campaigns (R ≈ 0.7).

- The HCHO/OA ratio is similar in both campaigns (≈0.32), but the observed R-values between HCHO and OA is higher in Europe than in the Asia (R ≈ 0.7 compared to ≈ 0.3).

By focusing on heavily polluted air masses sampled downwind in the city plumes, the ratios between the observed carbonaceous aerosols and the five trace gases change, and the R-values increase with O_3 for both BC and OA (R ≈ 0.5).

To assess the performance of atmospheric models with respect to the most relevant observed relationships, an air quality model ensemble is used to represent the current state of atmospheric modeling, consisting of two global and two regional simulations. The evaluation shows that these proportional relationships are not satisfactorily reproduced by the model ensemble. The relationships between BC and OA or between CO and BC are modeled with stronger correlations than the observed ones, and their higher ratios observed in Asia compared to Europe are not reproduced. Furthermore, the modeled HCHO/OA ratio is underestimated in the Asian campaign and overestimated in the European campaign.

This analysis of the proportional relationships between carbonaceous aerosols and trace gases implies that the observed relationships can be used to constrain models and improve anthropogenic emission inventories. In addition, it implies that information about the lower tropospheric concentration of carbonaceous aerosols can potentially be inferred from satellite retrievals of trace gases, particularly in the plumes from megacities.