Aerosol Composition Trends during 2000-2020: In depth insights from model predictions and multiple worldwide observation datasets

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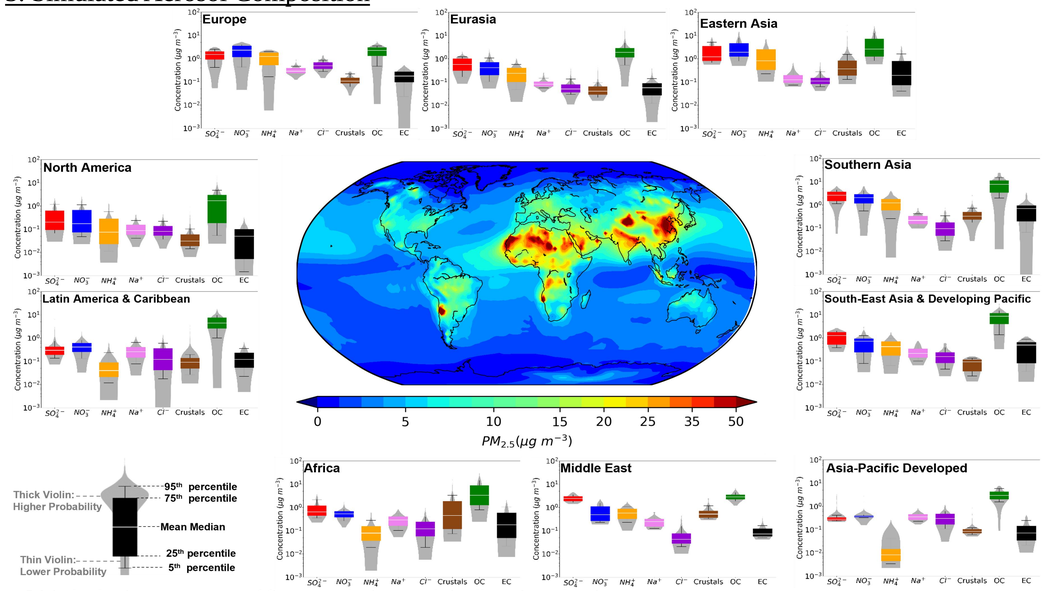
1. Abstract

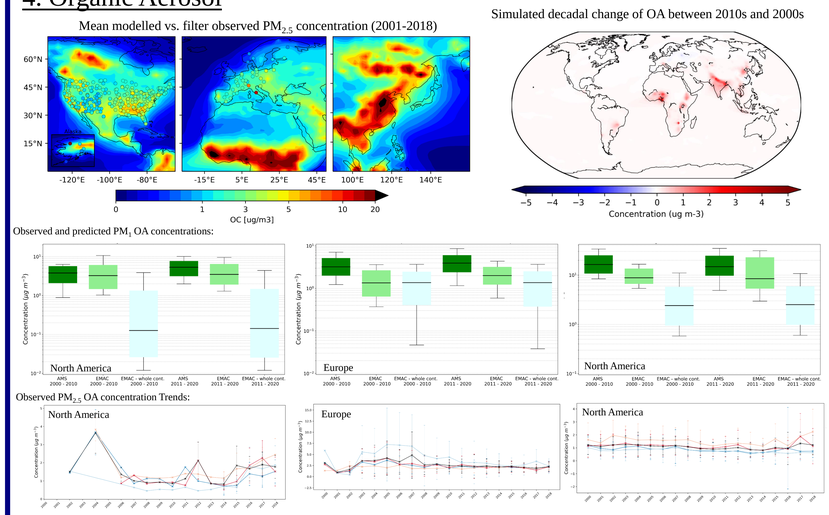
Atmospheric aerosol has a prime role on shaping the Earth's climate and deteriorating the air quality. Beside the number and mass concentrations of atmospheric aerosol, its chemical composition determines its aerosol-related climatic and health impacts. Emission trends since 2000 have been so drastic that aerosol composition has been unevenly altered in different parts of the world. Here, we use the comprehensive atmospheric chemistry-climate model EMAC to present 20-year global aerosol composition trends of fine aerosols in different regions of the planet. The large emission trends into our model are considered by employing the Copernicus Atmosphere Monitoring Service (CAMS) inventory for anthropogenic emissions. Model results are combined with a global observational dataset during the period 2000-2020 covering PM2.5 aerosol composition from regional routinely monitoring networks (e.g., EMEP in Europe) and PM1 aerosol composition from a comprehensive compilation of 744 aerosol mass spectrometry (AMS) observations during field campaigns conducted at 169 sites all around the world. A comprehensive analysis of model results and observations reveals valuable insights into the large spatiotemporal changes of atmospheric aerosol composition over different regions of the planet driven by recent changes in aerosol precursor emissions.

3. Simulated Aerosol Composition



- OA is the dominant component of fine atmospheric aerosols in all continental regions
- Sulphate follows the organic components in the aerosol composition over most regions
- Nitrate is the dominant inorganic component in Europe, Eastern Asia, Asia-Pacific, and Latin America

4. Organic Aerosol



- OA is captured well by the model in most cases; The model fails to reproduce the high OA concentrations over Europe during wintertime and over Eastern Asia during haze events.
- N. America: A negligible downward trend in rural and a slight positive trend in urban areas is observed. The positive trend is reproduced by the AMS and the model.
- Europe: Filter shows a slight yearly decline. The opposite trend can be observed AMS and the model.
- Eastern Asia: OA has been unevenly altered in different parts of the region

2. Aerosol Model Configuration

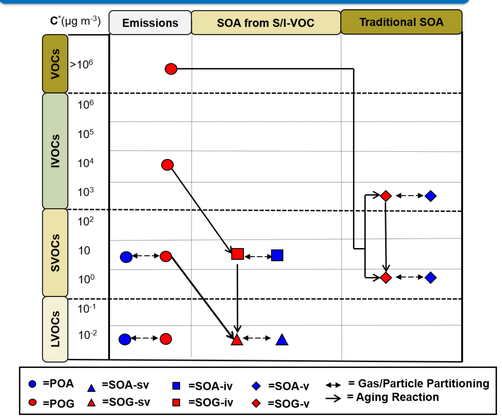
ISORROPIA-lite

K^+, Ca^{+2}, Mg^{+2}, NH_4^+, Na^+, SO_4^{-2}, NO_3^-, Cl^-, H_2O

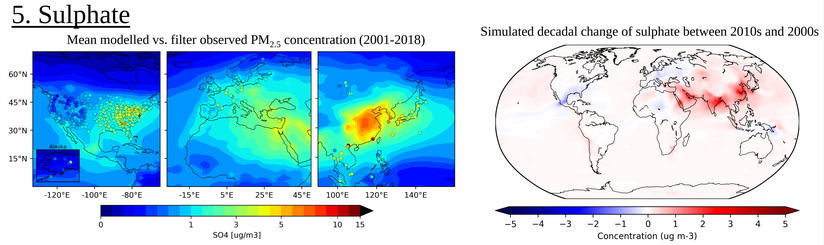
- The coefficients are taken from lookup tables
- Aerosols exist only as aqueous solutions even at low RH

ORACLE-lite

Minimizes the number of surrogate species used to describe OA formation from LVOC,SVOC,IVOC, and VOC

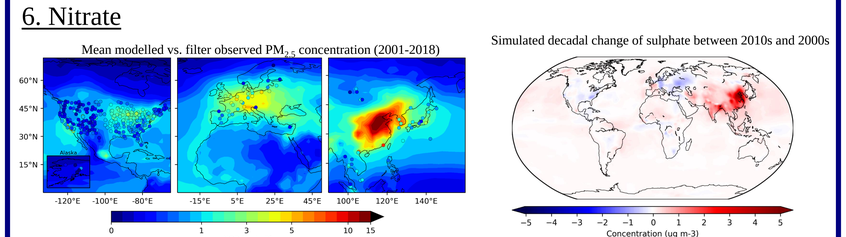


5. Sulphate



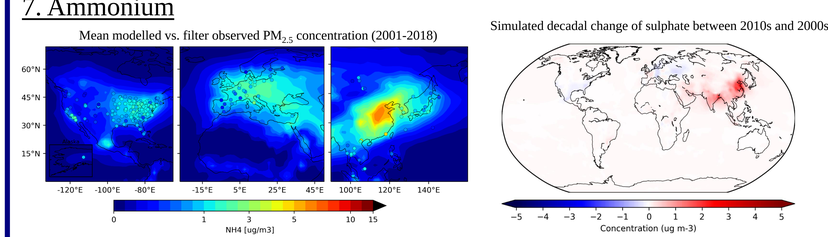
- The model performs well against filter-based observations, however, it undepredicts sulphate concentrations derived from AMS over Europe and Eastern Asia, especially during the first decade.
- N. America: Sulfate concentrations show a downward tendency (stronger close to urban locations).
- Europe: Sulfate strongly declines over the whole observational and modeled data.
- Eastern Asia: Simulated sulfate concentrations soared in Asia; however, this is not depicted in observational data over Eastern Asia since sulfate starts declining after 2015 when most AMS data has been collected.

6. Nitrate



- The model tends to overpredict the nitrate concentrations, mostly over the remote areas of North America
- N. America: $PM_{2.5}$ nitrate shows a decline during the past 20 years while both AMS and the model exhibit an upward trend.
- Europe: Simulated and observational data suggest a decline in nitrate concentrations. Nitrate increases in parts of western Europe, associated with the decrease of sulfate and increase of ammonium in the area.
- Eastern Asia: A slight upward trend is observed and modelled.

7. Ammonium



- EMAC represents ammonium concentrations very well with only a few underestimations over Eastern Asia.
- N. America: Filters show a strong reduction of ammonium concentrations while AMS and the model show only a slight decrease.
- Europe: Observations and the model suggest a downward trend in ammonium concentrations
- Eastern Asia: Meaningful concentrations changes have been observed.