SPECIAL PROJECT PROGRESS REPORT

All the following mandatory information needs to be provided. The length should *reflect the complexity and duration* of the project.

Reporting year	2025
Project Title:	FLEXPART transport simulations and inverse modelling of atmospheric constituents
Computer Project Account:	spatvojt
Principal Investigator(s):	Marina Dütsch
Affiliation:	University of Vienna – Department of Meteorology and Geophysics
Name of ECMWF scientist(s) collaborating to the project (if applicable)	……………………………………………………………… ………………………………………………………………
Start date of the project:	2024
Expected end date:	2026

Computer resources allocated/used for the current year and the previous one
(if applicable)
Please answer for all project resources

		Previous year		**Current year**	
		Allocated	Used	Allocated	Used
High Performance Computing Facility	(units)	2000000		2000000	
Data storage capacity	(Gbytes)	4000		5000	

Summary of project objectives (10 lines max)

The Lagrangian particle dispersion model FLEXPART (Stohl et al., 2005, Bakels et al., 2024) is run with ECMWF data to explore the dispersion and transport of various atmospheric constituents. The model is used with inversion techniques to enhance the knowledge about the emissions of many atmospheric compounds. This helps to get a better understanding of their impact on the Earth's climate system and air quality and to improve transport simulations of these substances. By performing domain-filling simulations the model is used to develop Lagrangian climatologies of heat and energy transport in the atmosphere and to perform case studies of extreme weather events.

Summary of problems encountered (10 lines max)

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Summary of plans for the continuation of the project (10 lines max)

We will continue the analyses on our research topics (see summary of results) and will try to finalize FLEXWEB, a web service where FLEXPART can be run in the cloud via a web interface.

List of publications/reports from the project with complete references

Bakels, L., Tatsii, D., Tipka, A., Thompson, R., Dütsch, M., Blaschek, M., ... & Stohl, A. (2024). FLEXPART version 11: Improved accuracy, efficiency, and flexibility. *Geoscientific Model Development*, 17(21), 7595-7627.

Bucci, S., Bakels, L., & Richon, C. (2024). Exploring the Transport Path of Oceanic Microplastics in the Atmosphere. Environmental Science & Technology, 58(32).

Tatsii, D., Gasparini, B., Evangelou, I., Bucci, S., & Stohl, A. (2025). Do Microplastics Contribute to the Total Number Concentration of Ice Nucleating Particles? Journal of Geophysical Research: Atmospheres, 130(2), Article e2024JD042827.

Summary of results

If submitted **during the first project year**, please summarise the results achieved during the period from the project start to June of the current year. A few paragraphs might be sufficient. If submitted **during the second project year**, this summary should be more detailed and cover the period from the project start. The length, at most 8 pages, should reflect the complexity of the project. Alternatively, it could be replaced by a short summary plus an existing scientific report on the project attached to this document. If submitted **during the third project year**, please summarise the results achieved during the period from July of the previous year to June of the current year. A few paragraphs might be sufficient.

1) Estimating Austria's greenhouse gas emissions using inverse modelling

Accurate estimates of greenhouse gas (GHG) emissions are essential to better understand the threats that climate change poses to various ecosystems and societies. The two gases that make up the largest proportion of all GHGs in the atmosphere are carbon dioxide (CO_2), which makes up about 76% of all GHGs, and methane (CH_4), which makes up about 16% (IPCC, 2014). In Austria, as on the global scale, the long-lived anthropogenic greenhouse gas that currently accounts for the largest share of Austrian greenhouse gas emissions is CO_2. According to the Austrian National Inventory Report (NIR) 2024, CO_2 hereby contributes to 84% of total GHG emissions in 2022 resulting primarily from combustion activities. The GHG with the second highest contribution is CH_4 with 8.9%. CH_4 emissions hereby arise predominately from livestock farming and waste disposal. Currently, the majority of GHG emission estimates in the NIR of different countries, including Austria, are obtained exclusively with the so-called *bottom-up* approach. Another method to estimate GHG emissions is the *top-down* approach which includes actual observations of atmospheric GHG mole fractions as well as atmospheric transport modelling. Top-down estimates can therefore be useful to verify or improve the bottom-up estimates by reducing uncertainties or providing an insight into unexpected and in many cases very significant errors (e.g., due to gas leaks, etc.). We examined the advantages and challenges of producing top-down estimates of Austrian GHG emissions using the stationary European observing network and currently available satellite data, with a focus on CO_2 and CH_4 emissions for the year 2022 in the framework of the GHG-KIT project.

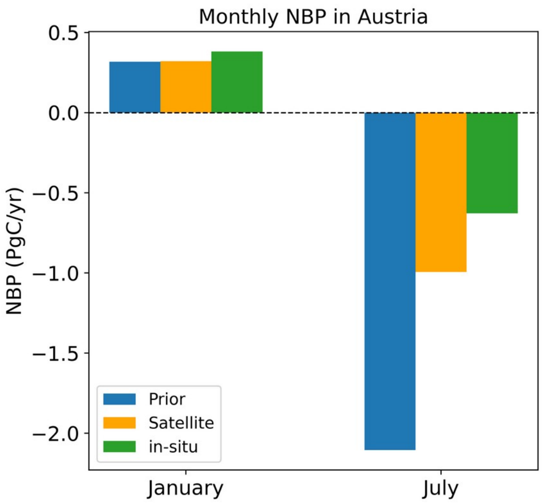

Figure 1: Monthly total NBP from Austria for January and July 2022 comparing a priori, satellite and in-situ based data.

For CO_2, the inversion optimizes net biome productivity (NBP), which is the sum of net ecosystem exchange (NEE) and CO_2 fluxes from disturbances such as biomass burning. Inversion using both in-situ and satellite observations suggests that the prior NBP over Austria is overestimated for the year 2022. In January, Austria acts as a source region of CO_2 since respiration dominates the photosynthetic uptake of CO_2 due to the cold temperatures, reduced sunlight, and limited plant activity. In January, flux corrections from satellite-based inversions are negligible. This is primarily due to the limited number of high-quality satellite observations over Austria during the winter months. The posterior flux from in-situ inversion shows slightly higher CO_2 emission from Austria in January. In July, Austria is a sink of CO_2 because vegetation maintains high photosynthetic activity, absorbing more CO_2 than is released through respiration. The sink capacity of Austria is reduced in July, as indicated by inversions using both in-situ and satellite data. The prior NEE is taken from the Vegetation Photosynthesis and Respiration Model (VPRM), which overestimates the CO_2 uptake. In July, the posterior NBP from in-situ inversion is –0.63 PgCyr-1 and that from satellite-based inversion is –0.99 PgCyr-1, both indicating reduced CO_2 absorption in Austria (Fig. 1). The magnitude of flux corrections from the satellite-based inversion is smaller than that from the in-situ inversion, due to the limited number of OCO-2 observations available during 2022. The temporal coverage of OCO-2 observations over Austria is insufficient to effectively constrain the fluxes. This highlights the importance of dense and consistent observational coverage for accurately constraining regional carbon fluxes through inverse modelling.

In the case of CH_4, the inversion results using the stationary observation data indicate that the prior emissions are underestimated for the year 2022. While the total prior, including anthropogenic and natural CH_4 sources, is about 700 Gg, the total posterior is about 910 Gg. According to Austria's NIR 2024, it was reported that 260 Gg of CH_4 were emitted from anthropogenic sources in Austria. This is lower than the estimated anthropogenic prior used for the inversion. However, it lies within the assigned uncertainty ranges. Except for the months of May and June, the posterior CH_4 emissions are higher than the prior in every month of the year 2022 (see Fig. 2). Significantly higher posterior CH_4 emissions were identified in February and March, indicating unusually high CH_4 emissions during these two months, most likely from anthropogenic sources.

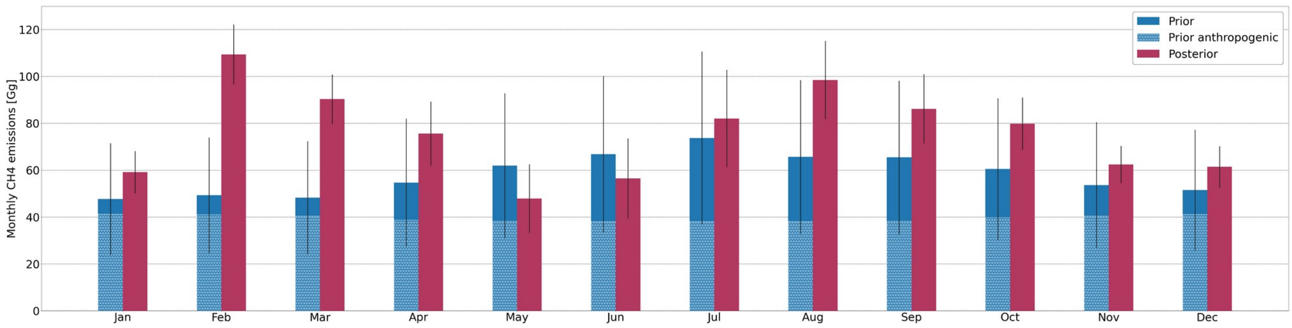

Figure 2: Monthly prior and posterior CH_4 emissions in Austria for the year 2022 (using in situ measurements for the inversion).

2) Estimating South Asian Methane emissions with TROPOMI-based inverse modeling

As one of the largest regional contributors to the global methane budget, South Asia acounts for about 26% of global anthropogenic CH_4 emissions between 2000 and 2017. However, the region's CH_4 sources and sinks remain highly uncertain due to the limitations of bottom-up inventories and the sparse ground-based observations.
To address these gaps, this study employs high-resolution satellite observations from TROPOMI, in combination with the FLEXPART Lagrangian transport model and the Bayesian inversion framework FLEXINVERT, to better constrain regional methane fluxes across South Asia. Despite the computational challenges of assimilating a large volume of satellite observations with a Lagrangian-based Bayesian inverse modeling system, the inversion framework demonstrated optimal performance in estimating methane fluxes over South Asia.

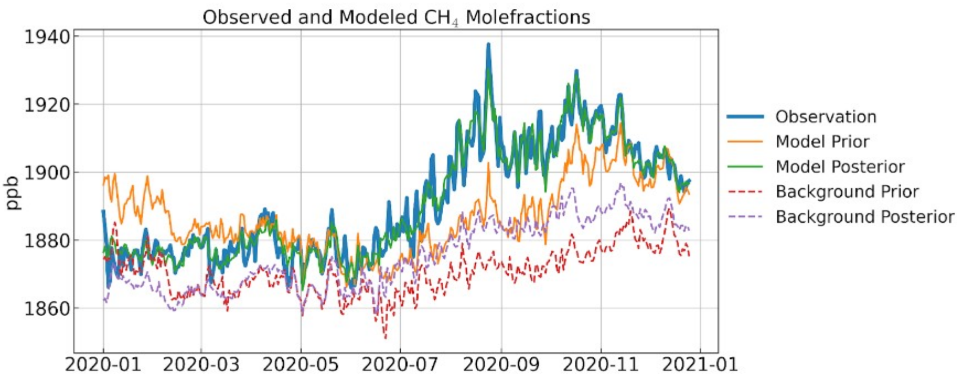

Figure 3: Time series of domain-averaged observed and modelled CH_4 concentrations for South Asia.

The results of this study highlight several important insights. The study reveals key limitations in existing methane emission inventories, particularly their inability to capture the seasonal variability of emissions (Fig. 3). The inventories overestimate the emissions from Jan to May and underestimate them from July to Dec. The spatial patterns are also inadequately represented, especially for emissions from agricultural and wetland sources, which dominate in many parts of South Asia (Fig. 4). Furthermore, the observed rise in methane levels during 2020 is likely attributed to increased biogenic emissions, driven by enhanced microbial activity under favorable environmental conditions.

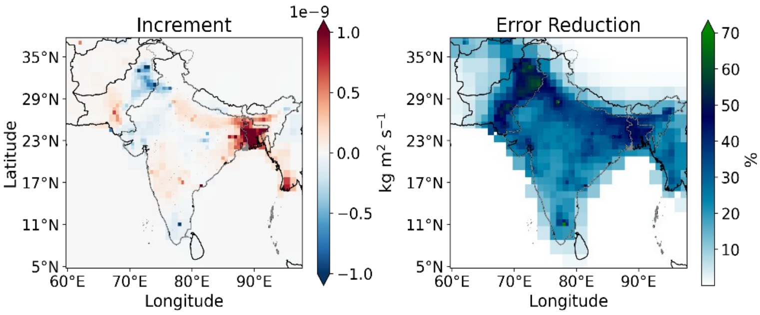

Figure 4: Spatial distribution of methane emission increments (posterior minus prior) across South Asia for the year 2020. Positive values indicate regions where emissions are underestimated in the prior inventories.

3) Sources of microplastics

Microplastics (MPs) are an emerging pollutant that appears to be ubiquitous in the atmosphere, usually with particle sizes comprised between nanometers and few tenths of micrometers. This has serious consequences, especially in terms of health impact when breathed, but also for its consequences over environmental contamination and possibly climatic impacts. Despite that, very little is still known about their actual sources in the atmosphere and the intensity of the fluxes in the air. It is also still not clear which type of sources are affecting the atmospheric concentration the most. In this study, we investigate the possible sources by using a Lagrangian approach. We start from datasets of MP atmospheric concentrations on different location, collected over long time series (order of few weeks and longer), to explore if any relationship can be found between the concentrations and the atmospheric transport variability. To track the atmospheric transport, we use FLEXPARTv11 backward trajectories starting from each data collection point, and using ERA5 global data at 0.5 degrees resolution as meteorological input data. We release 10,000 backtrajectories from a 3D box of 0.1° x 0.1° x 50m in a point centered around the measurement station position, released every 3 hours along the duration of each measurement campaign. The trajectories travel back in time for 45 days, a trade off between the typical lifetime of these particles in the atmosphere and the eventuality of getting long range transport events.

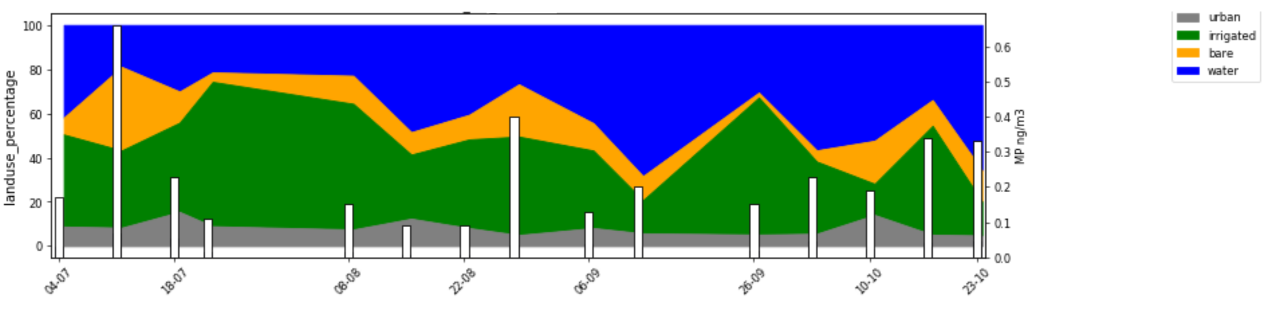

Figure 5: MP concentrations at Pic du Midi July to October 2017 (white bars) and percentage of simulated air mass influence from different surfaces, as classified from the global land cover (LC) products for the years 2017 in the framework of the Copernicus Climate Change Service (C3S) https://www.esa-landcover-cci.org/?q=node/199.

We show here an example of case study in one of the campaigns, from the data of Allen et al. 2021. This is a very interesting dataset, as it has been collected in the free troposphere (altitude of 2877 m), therefore indicative of the processes of long-range transport. They found concentrations of 0.09–0.66 MP particles/m3 over 4 months (Summer 2017) from the Pic du Midi Observatory, in France, for particles with diameter Dp <50 µm. Figure 5 reports the concentrations observed (white bars) vs the modelled percentage of influence of atmospheric transport from 4 different types of land use (Urban, Irrigated soils, Bare soils and Water). The choice has been made considering the possible plausible sources of MP emissions in the air: urban areas (due to direct human activities), agricultural fields (large use of plastic, as in mulching and hidden in sewage sludge fertilizers), desert regions (as dust has been shown to be sometimes associated to MP atmospheric transport) and water bodies (sea spray from polluted waters can inject MP in the atmosphere). It is difficult from this analysis to see any evident correlation between the type of landuse and the concentration of the particles. Sometimes the peaks are associated to an increase in arid soil influence, sometimes to water bodies influence and in once care with irrigated agricultural fields influence. A closer analysis has been performed considering also the possible emission scenarios, i.e. considering also the MP surface spatial distribution and not just the type of surface, focusing on specific sub-sectors (Bucci et al. 2024 for the ocean sources, Evangelou et al. 2024 for the dust sources and Tatsii et al 2025 for the urban sources). The analysis revealed that, among the various sectors, the dust-related one had the highest correlation with the MP concentration (54% vs values below 20% for the other emission sectors), see Figure 6. This suggests that, for this site, the majority of observed MP has been transported from desert dust-related sources. This is confirmed by the data at the site, which indicates the presence of mineral particles in the same days.

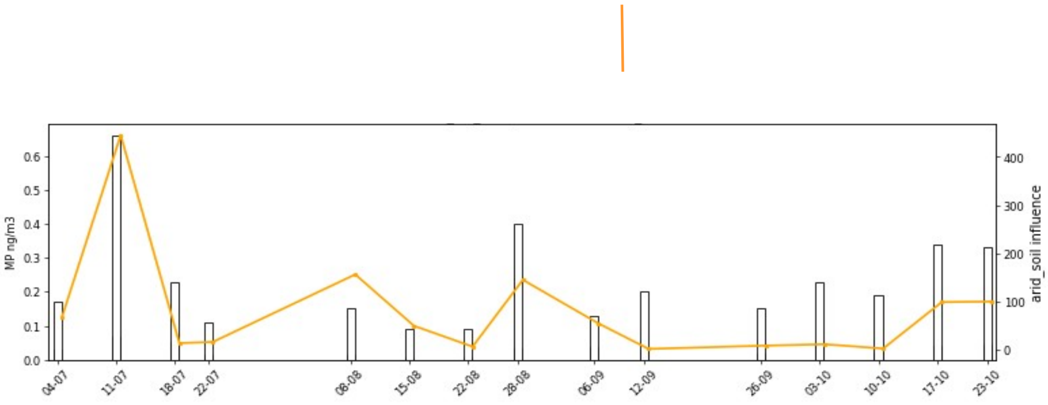

*Figure 6: MP concentration (white bars) and in orange the simulated influence of MP from arid soil (units based on artificial index, proportional to the possible MP emissions * probability of getting air masses from those regions).*

4) Evaluation of microplastic emission inventories

The atmospheric MP cycle is not well known. Available reported emissions differ by orders of magnitude based on the estimation method (top-down or bottom-up) and the particle size distribution assumed. The atmospheric MP cycle remains inadequately characterized in the current literature. Reported emissions exhibit significant variability, often differing by several orders of magnitude based on the estimation methodology employed (top-down or bottom-up) and the assumptions related to particle size distribution. Recently, the increasing number of atmospheric MP concentration measurements has given the opportunity to provide valuable insights into these emissions.
We employed the FLEXPART model in backward mode, from the locations of the measurements, using various wet scavenging scenarios (minimum, base, and maximum). The model was driven by the hourly meteorological re-analysis dataset ERA5 from the European Centre for Medium-Range Weather Forecasts (ECMWF), which is available at a resolution of 0.5° × 0.5°. The output from

FLEXPART was integrated with three emission inventories, two top-down approaches (TD-B and TD-E) and one bottom-up approach (BU), to compute the modeled MP concentrations.
Our findings reveal that all emission inventories generally overestimate the concentrations observed in situ (Fig. 7). The median modeled MP concentrations were found to be 2 to 4 orders of magnitude greater than the median measured concentrations.

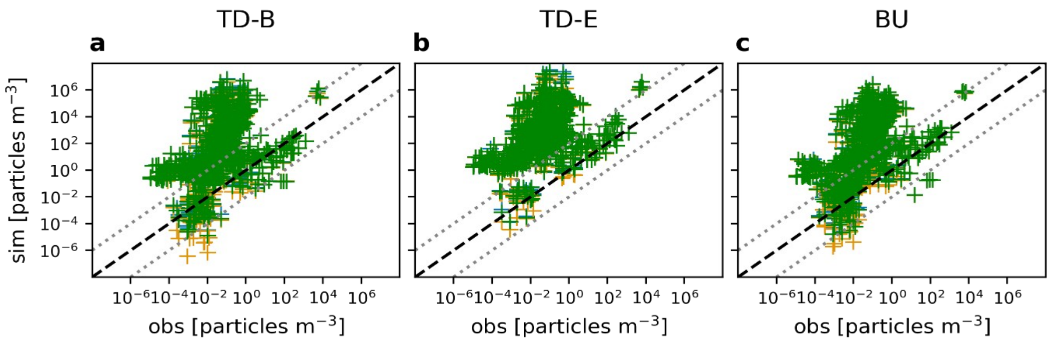

Figure 7. Scatter plots of observed (obs) and simulated (sim) atmospheric concentrations globally for the three emission inventories (TD-B, TD-E, BU). The green, blue, and orange crosses correspond to the base, minimum, and maximum scavenging cases, respectively. The bold dashed line corresponds to the 1:1 line, while the dotted lines correspond to the 100:1 and 1:100 lines.

5) Microplastics as ice nucleating particles

MPs can also be transported into clouds, where they may act as ice nucleating particles (INPs). However, MPs have not previously been considered as a source of INPs. We quantified the number concentrations of MPs related to road traffic and estimated their contribution to the total number of INPs. To do this, we estimated road traffic-related MP emissions from tyre and brake wear, road markings and polymer-modified bitumen. We then developed size distribution scenarios, estimated the airborne fractions for each size class and performed forward-in-time simulations of the atmospheric transport of MPs using a Lagrangian particle dispersion model FLEXPART. We then estimated the fraction of MPs acting as INPs and compared the obtained values with existing estimates of total INP number concentrations in mixed-phase (mineral dust and marine-sourced aerosols) and cirrus (mineral dust) clouds.
We suggested two emission scenarios of road traffic-related MPs: 259 ± 9 kt/year for the MIN scenario and 1005 ± 139 kt/year for the MAX scenario. Under MAX scenario, we found that ice-active MPs could account for between 0.1% and over 40% of the total INP number in immersion freezing conditions in the tropics (Fig. 8). For cirrus conditions, their contribution could be as high as 7% over the tropical Pacific and 20% over East Antarctica. Therefore, in regions where other effective INPs are scarce, concentrations of ice-active MPs may be sufficient to trigger the heterogeneous nucleation of ice crystals in mixed-phase clouds or cirrus. Thus, MPs may be able to substantially affect cloud formation. These findings provide new insights into the potential role of MPs as INPs, highlighting their possible impact on cloud formation and their properties.

6) Lagrangian re-analysis dataset

We have finished the creation of a Lagrangian re-analysis, based on the ERA5 dataset from ECMWF (Hersbach et al., 2020), on a one-hourly basis. We divided the full time range in 12 periods with a one year overlap. We ran our domain-filling transport model simulation with the Lagrangian particle dispersion model FLEXPARTv11, using six million particles. This dataset is suitable e.g. for establishing transport climatologies and global statistics. It is publicly available at https://data.eodc.eu/collections/LARA/, and a paper describing the dataset is currently in review for Earth System Science Data.

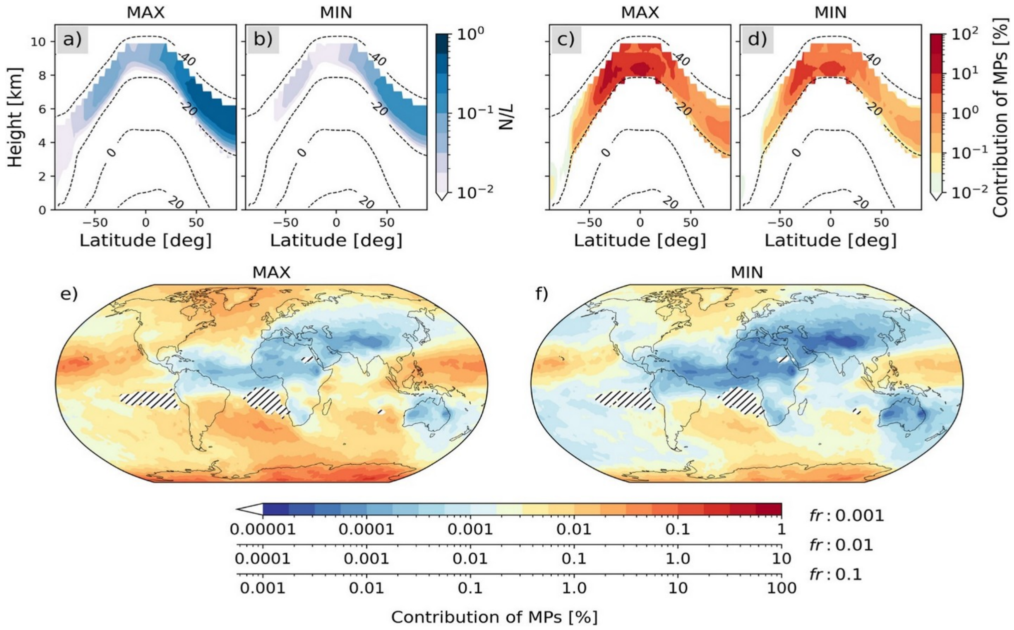

Figure 8. Annual mean of microplastic INP number concentration (a and b) and contribution of the road traffic-related MPs to the total number concentration of INPs in mixed-phase regime as modeled by Herbert et al. (2025) (c and d) and in cirrus cloud conditions as modeled by Beer et al. (2022) (e and f). Number concentration is expressed in N/L (particles per liter). Panels (a, c, and e) and panels (b, d, and f) show MAX and MIN scenarios, respectively. Black dashed lines are climatological mean isotherms. The three colorbar tick labels under panels (e and f) represent the value ranges when assuming different fractions of cirrus-active INPs from MPs (fr), corresponding to 0.001, 0.01, and 0.1. No cirrus clouds are present in hatched areas.

7) FLEXWEB

We are developing a web service, called FLEXWEB, where FLEXPART can be run in the cloud via a web interface. FLEXWEB uses ERA5 data as input for the meteorological fields and gives users worldwide access to FLEXPART without the need for installation on a local HPC system or downloading meteorological input data. A first version of the web service is already running on our local cluster (https://flexweb.wolke.img.univie.ac.at/), and we are currently working on transferring it to a Kubernetes-based service on the European Weather Cloud (EWC).

As part of this process, we have encountered several challenges. We managed to deploy a Kubernetes Cluster and contributed a bug fix for automated deployment with the help of EWC's excellent support team. Upon successfully deploying a Kubernetes cluster on the EWC, we identified key areas for improvement in the application's design and functionality. Specifically, our web service requires further optimization to enable seamless deployment, scalability, and efficient resource utilization. One critical bottleneck is the manual scaling of worker nodes based on job demand, which necessitates a more robust dynamic scaling mechanism. We aim to redesign our dedicated containers to incorporate a different metric that enables optimized resource allocation. Another area for enhancement is the plotting of simulation results, which currently consumes excessive time and resources. To address this, we plan to re-engineer the application's architecture to improve performance and reduce runtime. Additionally, as our initial version did not include direct data retrieval from MARS, we will need to develop a cache server to facilitate the delivery of input data to worker nodes, enabling faster and more efficient processing.

References

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