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	2023
Project Title:	FLEXPART energy transport simulations and inverse modelling of atmospheric constituents
Computer Project Account:	spatvojt
Principal Investigator(s):	Martin Vojta
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:	2021
Expected end date:	2023

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)	20000		30000	

Summary of project objectives (10 lines max)

The Lagrangian particle dispersion model FLEXPART (Stohl et al., 2005, Pisso et al., 2019) is run on 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 better understand their impact on the Earth's climate system and air quality and 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 results and plans for the continuation of the project

1) Lagrangian re-analysis Dataset

The second phase of the Lagrangian re-analysis has been completed. This re-analysis dataset is created by using domain-filling transport model simulations with a Lagrangian particle dispersion model (FLEXPART). This dataset makes it possible, for example, to produce forward and backward simulations of particle locations, Lagrangian transport climatologies, and global statistics of atmospheric transport (see previous report).
This newest set consists of 6 million particles, advected with the newest and improved version of FLEXPART (Bakels et al. in prep.). In this new version, the accuracy is improved due to fewer interpolations, and the initial particle positions are more homogeneously distributed. In addition, the set is run over a longer time period, now ranging from 1959 to 2022, and includes both time-averaged and instantaneous hourly particle information.

By using this Lagrangian Reanalyses Dataset (LARA), the energy export from the Equatorial Pacific, and how it is affected by the El-Niño-Southern-Oscillation (ENSO) is detected. Therefore, only particles that have been in the Nino3.4 and Nino3 region and below 1 km were selected and followed 20 days forward in time. We then determine the mass fraction r of the transported tagged air relative to its environment, the total air, present in a volume. We further determine how different the transported tagged air is relative to the untagged environmental air in potential temperature and specific humidity. By using the Nino3.4 Index, we consider how these impacts of the transported tagged air are changing with ENSO. Based on that we found significant anomalies in moisture and heat compared to its environment.

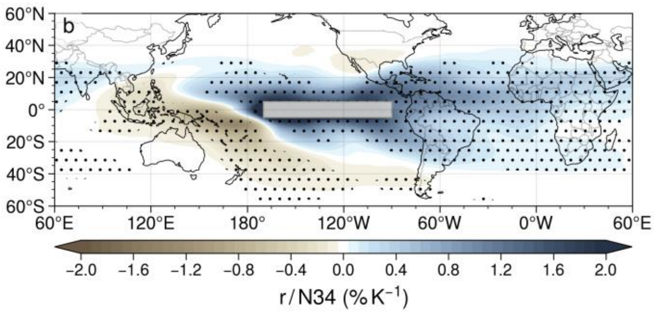

Figure 1: Mass transport from the Equatorial Pacific for DJF. Slope of the regression of the mass fraction r with the Nino3.4 Index (N34).

Furthermore, the Lagrangian Reanalyses Dataset is quite useful in order to study extreme events. For example, we studied the extreme heat wave over Western Canada in June 2021. Therefore, particles from the target region are followed backward in time, in order to identify where they are coming from and how their meteorological properties are changing along the transport. We did additional FLEXPART simulations for the single extreme event, with ERA5 as input data.

In the future, we are planning to do the simulation with the Operational data from ECWMF.

2) Inverese modeling of F-Gases

Inverse modeling provides a powerful tool to verify national greenhouse gas (GHG) emission inventories by using atmospheric observations. Our inversions are based on Lagrangian Particle Dispersion Model simulations by FLEXPART, which is run on the ECMWF ERA5 data. Here, virtual particles are released from observation sites and traced backwards in time to establish a relationship between atmospheric concentrations and emission sources within the simulation period. The fact, that this simulation period is limited due to computational costs, raises two essential questions: (i) How to best define a baseline, that accounts for all emissions that occur prior to the simulation period? (ii) Which period length should be chosen for the backward simulation?

We have shown that often-used statistical baseline methods have large problems and present a superior global-distribution-based (GDB) approach, that is consistent with the backward-simulation period, accounts for meteorological variability, and leads to inversion results that agree well with known global emission estimates. Our results further show, that longer backward-simulation periods beyond the often used 5 to 10 days increase the correlation between modeled and observed concentrations, and lead to more robust inversion results. Furthermore, they help to better constrain emissions in regions poorly covered by the observation network.

Based on these methodological results, we perform inversions for sulfur hexafluoride (SF_6) - the most potent GHG regulated under the Kyoto Protocol with an estimated atmospheric lifetime of 3200 years.

The inversions are based on 50-days backward simulations, in-situ and flask measurements from various observation networks, and the GDB baseline method, to achieve global and European SF_6 emissions from 2005 to the present. Our recent inverse model results (Figure 2) show that European SF_6 emissions have been overall decreasing between 2005 and 2015, although the inversion results are higher than the reported emissions.

Figure 2: European SF_6 emissions between 2005 and 2015. The green bars show the reported emissions (a priori) and the red bars show the inversion results (a posteriori).

In the future, we will also perform time series of global and regional emissions of other F-gas species, such as HFC_134a, HFC-23, HFC-125, HFC-143a, and HFC-32.

3) Inverse modeling of methane over India using satellite column observations

The source-sink estimation of greenhouse gases (GHG) and the accurate quantification of their flux distributions are major scientific challenges of our times. In order to constrain the GHG budget and facilitate a reduction in uncertainty, atmospheric GHG observations are used together with numerical models, popularly known as top-down methods. There are, however, significant gaps in the GHG in-situ observation network, making it troublesome to constrain their budget. Fortunately, in this era of satellite remote sensing, it is possible to use high-resolution data from satellites to estimate the GHG budget.

Our work aims to constrain methane fluxes over the Indian region which are not well understood and poorly constrained due to the lack of sufficient in-situ observations. The study uses column observations (Figure 3) from the Tropomi instrument onboard the Sentinel-5p satellite for constraining the Indian methane budget. This is achieved by using FLEXPART running in backward mode to produce the column sensitivities of the molar mixing ratios at the receptor locations to the fluxes, which is then used along with the Bayesian inversion system FLEXINVERT to obtain *a posteriori* emissions. For this purpose, various prior fluxes of methane are prepared including fluxes from fossil fuel burning, biogenic fluxes, fire fluxes, biomass burning and wetland fluxes. Column background mole fractions are calculated using a satellite averaging kernel and *a priori* information which is then used to compute the modeled methane column concentrations. The new developments in the FLEXINVERT model for satellite data assimilation are first tested for stability and the results will be compared with the limited ground-based observations available.

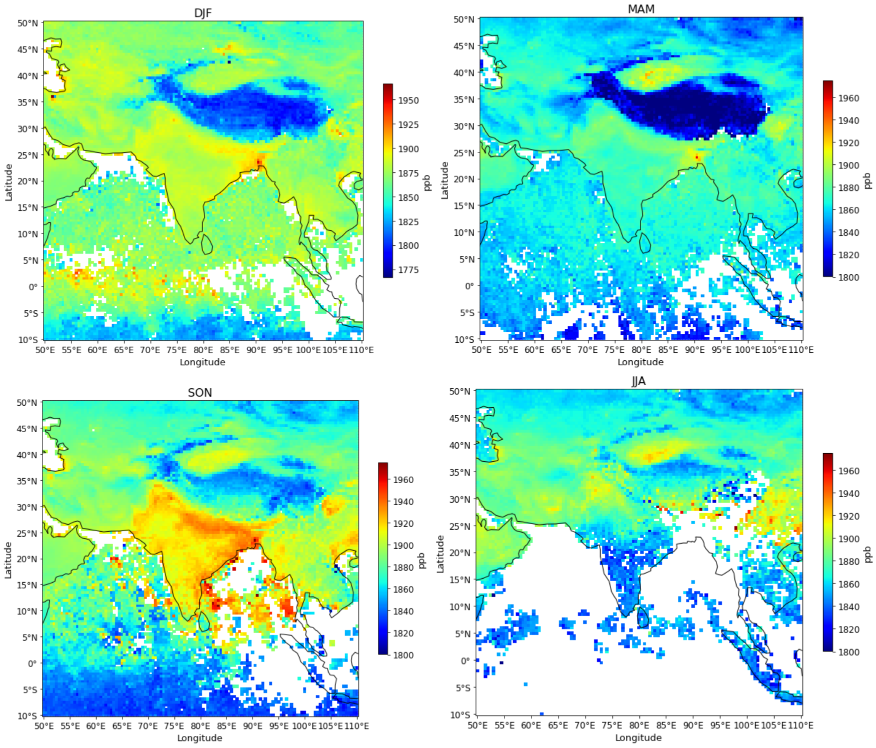

Figure 3: Methane column mole fractions over the Indian region averaged for the four seasons, from the Tropomi retrievals

4) <u>Interpretation of ice core data</u>

We are collaborating with scientists all over the world by helping to interpret deposition of various species found in ice core records. It is essential to understand the atmospheric pathways the deposited particles took, in order to be able to identify potential emission source regions. For this, we run global FLEXPART simulations driven by ERA5 re-analysis data at a horizontal and temporal resolution of 0.5° and 1 hour, respectively. FLEXPART is used in backward mode, releasing virtual particles and sending them backward in time from individual ice cores sites to identify potential emission source regions for each ice core site. However, FLEXPART is also used in forward mode, releasing particles from known or suspected emission sources to identify the most promising ice core sites at which most deposition might have occurred.

In this field of research, we have recently been involved in studies investigating:

- European Phosphorous emissions from Alpine ice cores.
- Global Sulfur dioxide emissions of the last century from an array of ice cores via inversion.
- The 20th-century Thallium emissions in Western Europe as recorded in an ice core extracted at the Col du Dome site in the French Alps.
- Sources of black carbon recorded in an array of Northern mid- and high-latitude ice cores from 1850 to 2000.
- Early Andean, Spanish Colonial, and Industrial-era mining emissions recorded in several Antarctic ice cores.
- The decoupling of Canadian forest dynamics from climate following European settlement by analyzing pollen preserved in two Greenlandic ice cores.
- The consistent histories of anthropogenic Western European air pollution preserved in four Alpine ice cores from 1750 to 2015.
- The increased black carbon emissions in the Southern Hemisphere following the extent of human settlements, combining low-latitude lake sediments and Antarctic ice core records.
- Changes of ammonia and nitrogen oxide emissions in South-Eastern Europe inferred from an Elbrus (Caucasus, Russia) ice core record (1774-2009 CE).

5) Atmospheric microplastic emissions from the Ocean

In the context of oceanic exchanges with the atmosphere, a new pollution problem has begun to emerge: the possible injection of microplastic particles from the sea surface into the atmosphere. Correctly assessing the emissions and dispersion of plastic particles is an increasingly important question, as a growing number of studies have shown that microplastics transported in the atmosphere have harmful effects on the ecosystem and human health.

The presence of microplastics in the ocean has long been recognized as a pollution threat to the marine environment, and recent studies have indicated that processes such as wave breaking and bursting bubbles can ultimately inject marine microplastics into the atmosphere with sea spray. Adopting a bottom-up approach to estimate the fluxes at the ocean-atmosphere interface, we start with the concentrations of microplastics at the ocean surface simulated general circulation model NEMO-PISCES (Nucleus for European Modelling of the Ocean, Pelagic Interaction Scheme for Carbon and Ecosystem Studies, C. Richon et al 2022) and apply a spray emission scheme (Grythe et al 2017) to estimate the emission fluxes from the sea surface. The emissions are then introduced into the FLEXPART atmospheric Lagrangian model (Pisso et al 2019) in order to quantify the impact of microplastics on the atmospheric distribution and their redistribution over the ocean and land (see Figure 4). This work led to an initial global representation of the distribution of microplastics in the atmosphere, associated with their oceanic sources. The associated article is currently being prepared.

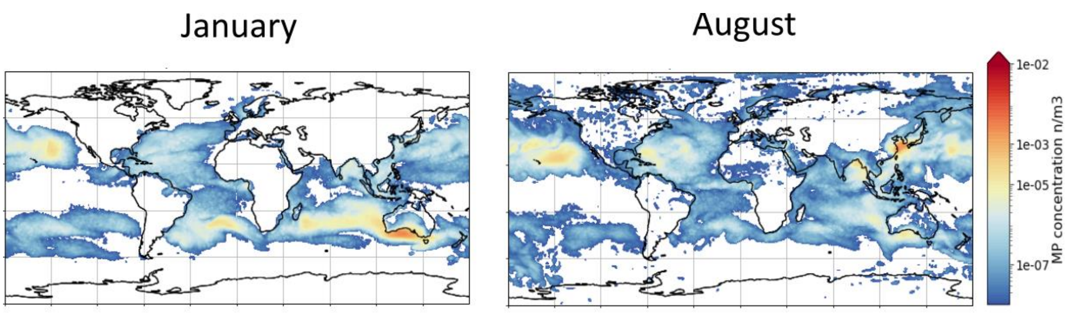

Figure 4: Monthly average of simulated particle counts or marine MP in the lowest km of the atmosphere. The values are representative of year 2014.

6) Investigation of the shape effect on atmospheric particles transport

Airborne microplastic particles can potentially have significant effects on human health and the environment in areas where they are deposited. Therefore, it is important to gain more knowledge about the mechanisms involved in their transportation and removal. This knowledge will enable the investigation of their distribution in situ, including their transport to remote regions. Currently, most atmospheric transport models consider particles as perfect spheres, neglecting the impact of non-spherical particles, which experience greater drag in the atmosphere. This increased drag leads to a reduction in their settling velocity and consequently prolongs their residence time in the atmosphere. As a result, there can be significant discrepancies between model predictions and ground-based measurements. To address this issue, our current research focuses on studying the gravitational settling of microplastic fibers, which are the predominant shape of microplastics (Rebelein et al., 2021). By doing so, we aim to enhance the accuracy of atmospheric transport models by eliminating uncertainties associated with particle shape, thereby improving simulations of atmospheric concentrations and deposition patterns.
In our study, we have incorporated a shape correction factor, developed by Bagheri and Bonadonna in 2016, into the gravitational settling scheme of the Lagrangian transport model FLEXPART (Pisso et al., 2019). The scheme was tuned according to experiments on the gravitational settling of fibers in air. For our simulations using FLEXPART, we utilize wind fields from ERA5 reanalysis data. To assess the model's sensitivity to the shape correction, we calculate average atmospheric transport distances and residence times for particles of various sizes and shapes, assuming particle emissions in different climatic regions and at different release

heights. In addition, we used a realistic scenario of land-based microplastic emissions (Evangeliou et al., 2022), disaggregated according to the population density data, and explored the resulting global atmospheric concentration and deposition patterns for spheres and fibers. We consider this simulation as a realistic approximation of global atmospheric microplastic transport for our purpose of exploring the sensitivity of the simulated transport to the particle shape. Four simulations were performed, in which it was assumed that all microplastic emissions are either spheres with a diameter of 75 µm, or cylinders of the same volume with AR values of 20, 50, and 100. The resulting deposition and mass concentration fields are shown in Figure 5.

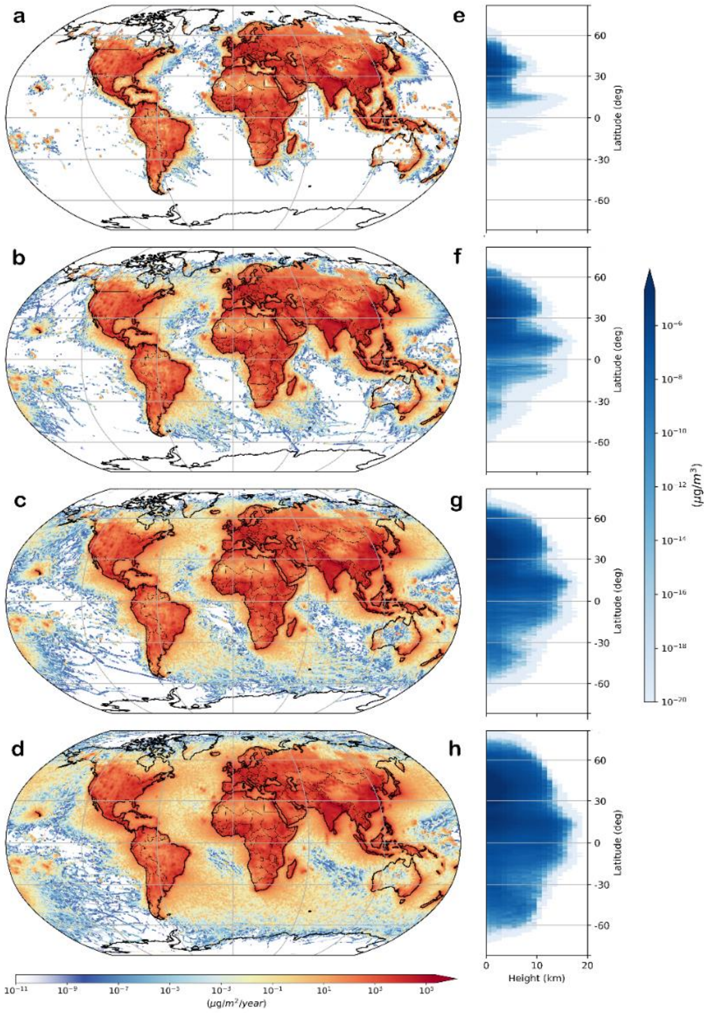

Figure 5: Shape dependence of microplastic particle deposition and vertical transport. Shown is the simulated total deposition in 2018 for spheres (a) and straight cylindric fibers with identical volume and AR=20 (b), AR=50 (c), and AR=100 (d) and the zonal median value of mass concentration for spheres (e) and straight cylindric fibers with identical volume and AR=20 (f), AR=50 (g), and AR=100 (h). Emissions are based on the population density map

Currently, we are collecting ERA5 reanalysis data on snow cover to conduct FLEXPART backward simulations to estimate possible sources of microplastics in remote areas of Siberia, Russia.

7) Nudged simulations with iCESM

Numerical models are useful for interpreting measurements of stable water isotopes, but the complex processes involving isotopic fractionation are often poorly represented. One example is evaporation from the ocean. In addition to equilibrium fractionation, it involves nonequilibrium fractionation if the relative humidity above the ocean is less than 100%. Many models parameterize non-equilibrium fractionation during evaporation from the ocean using the wind-speed dependent formulation of Merlivat & Jouzel (1979) even though it has been shown that the resulting fractionation factors do not agree with observations (e.g., Pfahl & Wernli, 2009; Bonne et al., 2019). We developed a new theoretical framework for parameterizing nonequilibrium fractionation during evaporation from the ocean, which accounts for waves in the momentum flux equation and is more in line with observations. The resulting fractionation factors were implemented in the isotope-enabled Community Earth System Model (iCESM; Brady et al., 2019) and tested by running simulations nudged to ERA5 reanalysis data (U, V, PS). Figure 6 shows the difference in vapor deuterium excess (δD-8·δ18O) between a simulation using the new parameterization and a simulation using the parameterization by Merlivat & Jouzel (1979). The new parameterization leads to higher deuterium excess, especially in regions with high wind speed. In a next step, the simulated values will be compared with measurements from the Antarctic Circumnavigation Expedition (ACE), which took place in 2017.

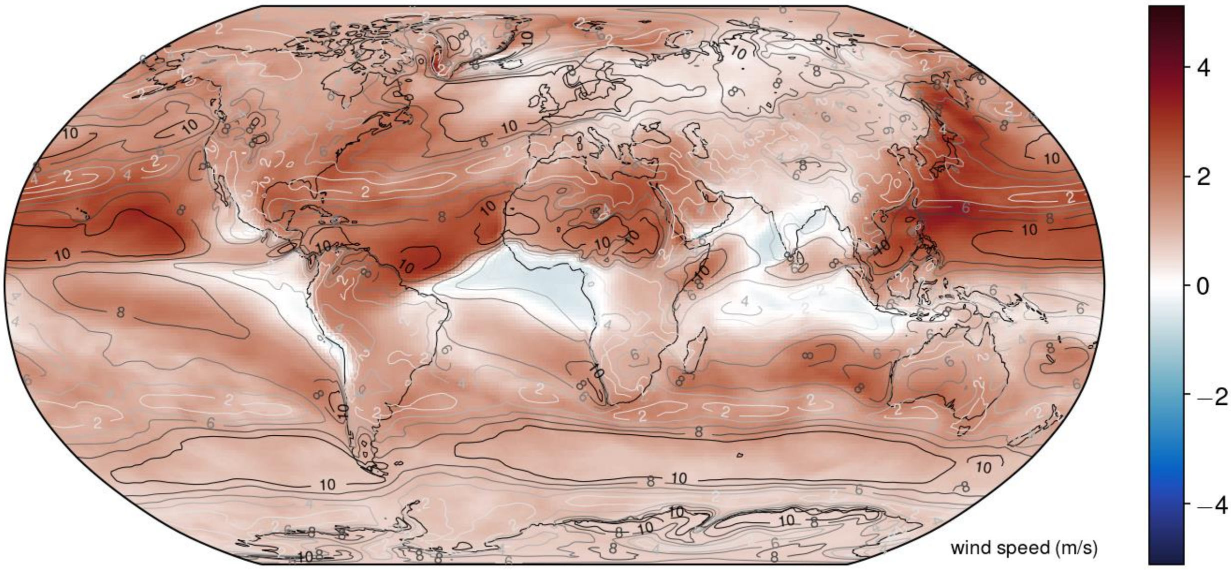

Figure 6: Difference in deuterium excess in vapor on the lowest model level in iCESM for January 2020 between a nudged simulation using the new parameterization of nonequilibrium fractionation during evaporation from the ocean and a nudged simulation using the parameterization of Merlivat & Jouzel (1979). The contour lines show wind speed in m/s.

List of publications/reports from the project with complete references

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