A03 Aerosol nucleation in the upper troposphere

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Motivation

Aerosol nucleation in the upper troposphere is an important source of cloud condensation nuclei (CCN). It is crucial to study the chemical composition and formation processes of nucleation mode particles (NMPs) to enhance our understand-ding of aerosol-cloud interactions and its role for climate.

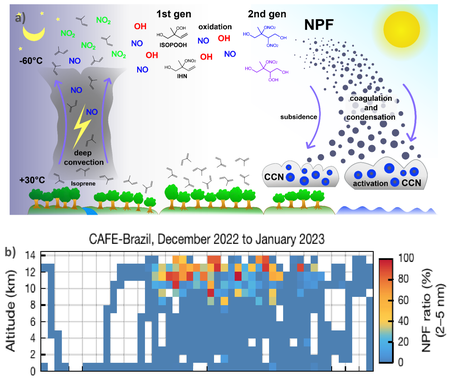

Figure 1: (a) Nucleation mechanism, (b) Occurence of NPF as function of altitude and time-of-day, [Curtius et al., Nature, 2024].

Objectives:

- Investigate precursor gases in the tropical and extratropical upper troposphere (UT) that drive new particle formation (NPF) by using a CI-APi-TOF onboard the research aircraft HALO.
- Analyze the amount of transported air masses and trace substances by convection with a hierarchy of models from the global to the local scale for CAFE-Brazil and globally.
- Analyze temporal trends of convective transport characteristics.
- Investigate aerosol growth processes from nucleation events to larger aerosol sizes due to coagulation and/or condensation.
- Examine abundance and distribution of NMPs in the extratropical UTLS with the newly designed multi-channel particle counter during the TPEx campaigns in northern Germany.

Collaborations within TPChange

A01 Influence of precursor gases (SO_2) on UTLS composition.

A04 Joint measurements of aerosol concentration, composition, size distribution during aircraft campaigns.

A05 Organic aerosol composition in the UTLS.

B01 Model simulations for the TPEx I and II campaigns.

B02 Aerosol microphysical modelling for balloon-borne aerosol observations.

C06 Transport and mixing of aerosols; growth to CCN.

C08 Convective transport on different scales and resolutions.

Z03 Data exchange with groups that participated in TPEx, aerosol modelling developments.

Results from phase I

CAFE-Brazil & CAFE-Pacific

We showed that isoprene (ip), emitted by tropical forests, is transported into UT by deep convection. At low temperatures (~210 K) the ip reacts to form oxygenated organic compounds (ip-OOMs). Through growth processes these can become CCN and contribute to cloud formation in the tropical troposphere.

Oxygenated organic compounds produced from Isoprene drive aerosol nucleation in the tropical UT.

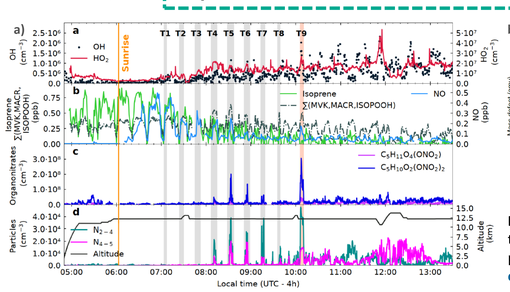

Figure 2: NPF events during CAFE-Brazil. (a) time series of key species and (b) mass-defect plot during strongest NPF event (T9), [Curtius et al., Nature, 2024]

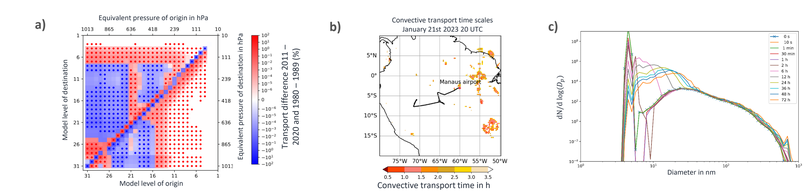

Figure 3: (a) Changes in the tropical convective transport due to warming [Jeske and Tost, ACP, 2025, preprint]. (b) convective transport times based on ERA5. (c) Aerosol size distribution development after NPF from microphysical modelling.

- Deepening of parameterized convective transport in the global and tropical trend using a convective exchange matrix.
- Short convective transport times (fast transport) during the CAFE-Brazil campaign and fast growth of aerosol particles after NPF.

mc-CPC for TPEx

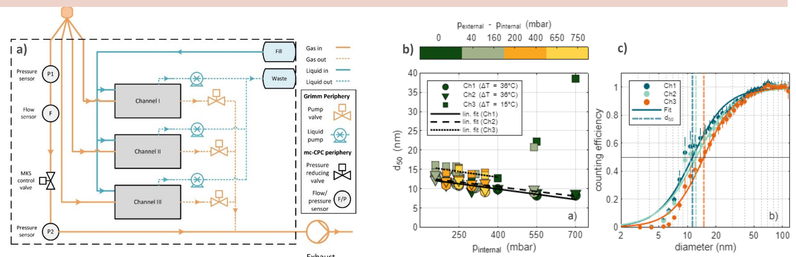

Figure 4: (a) Flow schematic of the mc-CPC, (b) CPC cutotffs as function of p_{CPC} and Δp, (c) counting efficiency and cutoffs of CPC channels, [Richter et al., AMT, 2025, submitted].

- Characterization of mc-CPC at different p_{CPC} and $p_{external}$.
- Operation during TPEx in northern Germany showing potential NPF events and aerosol concentrations $N_{11\text{-}15nm}$ up to 10,000 cm^{-3}.

Research plan phase II

Main goal: Identify chemical NPF precursors in the extratropical UT, examine reaction pathways & formation mechanisms and quantify rates of nucleation and growth.

CONTANGO-FIRE aircraft campaign

- Operation of a bipolar CI-APiTOF mass spectrometer, which can measure quasi-simultaneously negative and positive ions, and of a multi-channel Condensation Particle Counter (mc-CPC).
- Investigate precursor transport into UTLS by probing deep convective outflow.
- Perform high resolution simulations (global to local scale) based on MECO(n) simulations with comprehensive aerosol (NPF, microphysics and chemical composition) and gas phase (precursor chemistry) processes and detailed analysis of convective lifting, scavenging and transport.
- Support the flight campaign with forecasting and now-casting products for convection, clouds, and biomass burning, as well as chemical forecasting.

Figure 5: HALO mission CONTANGO-FIRE will be conducted from Mendoza, Agentina to study NPF in the subtropical UT.

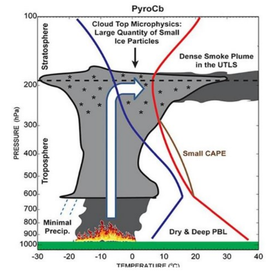

Figure 6: HALO mission CONTANGO-FIRE will also study effect of biomass burning emissions for UTLS region.

TPEx II aircraft campaign

- Operation of the mc-CPC that was designed, constructed, calibrated and operated during TPChange phase I.
- Investigate the abdundance of nucleation mode particles and the frequency of nucleation events over Europe.
- Accompanying modelling in cooperation with B01 & B02.

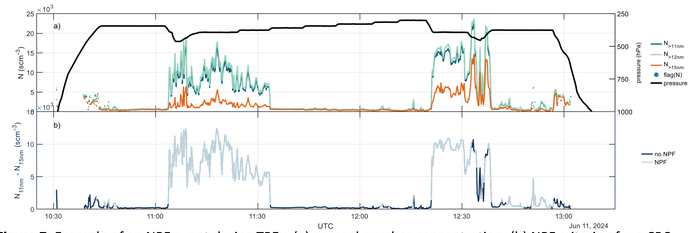

Figure 7: Example of an NPF event during TPEx, (a) aerosol number concentration, (b) NPF criteria of mc-CPC.