Using Trace Gas Measurements as a Transport Diagnostic for Tropical Convection: A Combined Model and Observation Study

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Introduction & Objectives

Enhance Understanding of Convective Transport:
Utilize VOC measurements from the CONTRAST field campaign.
Use tropospheric transit time distributions (TTDs) from the Whole Atmosphere Community Climate Model (WACCM).
Integration of Data:
Combine observation-based TTDs with model simulations.
Evaluate the representation of convective transport in global models.
Analysis Focus:
Compare model-derived lifetimes and TTDs with observational data.
Assess the model's ability to capture spatial and temporal variabilities.
Sensitivity Exploration:
Examine the sensitivity of modeled TTDs to different VOC lifetime definitions and source region treatments.
Uncertainty Quantification:
Quantify uncertainties to evaluate the accuracy of TTDs.
Highlight the importance of integrating observational data with simulations in assessing convective transport in chemistry-climate models.

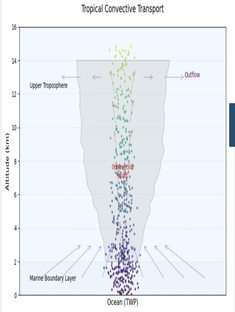


Figure 1. *Tropical convection transports surface-emitted VOCs to higher altitudes. This schematic shows air movement through the boundary layer into the upper troposphere, where outflow redistributes these compounds horizontally.*

CONTRAST Data:

CONvective TRansport of Active Species in the Tropics Campaign:
Conducted from January to February 2014 over the tropical western Pacific, this campaign explored how tropical deep convection affects atmospheric composition.
In-situ Measurements via Trace Organic Gas Analyzer (TOGA) and Advanced Whole Air Sampler (AWAS):
Utilized to gather detailed atmospheric data.
Vertical Profiles:
Nearly 100 profiles collected from convective systems, providing extensive sampling across the upper troposphere (UT) and boundary layer (BL).
Chemical Species Sampling:
Captured a wide range of chemical species with varying lifetimes, essential for studying vertical transport and the redistribution of trace gases.

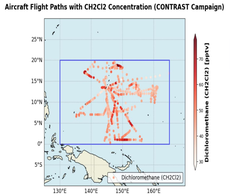


Figure 2. *Flight paths during the CONTRAST campaign, covering the TWP region where deep convection drives VOC transport from the boundary layer to the upper troposphere.*

WACCM Output

Whole Atmosphere Community Climate Model (WACCM)
Output: Provides daily averaged and flight track data.
Parameters Covered: Includes meteorological data, trace gas concentrations, and chemical loss rates.
Species Count: Focuses on 26 different chemical species.
Simulation Details: WACCM simulates the transport of these species throughout the atmosphere.
Tracer Dynamics: Tracers are emitted from the surface, with decay rates determined by their photochemical lifetimes.

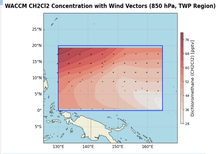


Figure 3. *WACCM Simulated CH2Cl2 Concentration and Wind Patterns in the TWP Region at BL.*

Data Segmentation for Atmospheric Analysis

Boundary Layer (BL) - 0 to 2 km: Essential for investigating surface-related chemical activities and emissions.
Upper Troposphere (UT) - 12 to 14 km: Targets the analysis of chemical processes at higher altitudes influenced by convective activities.
Whole Troposphere - 0 to 14 km: Covers the entire vertical span of the troposphere to assess comprehensive transport and mixing of atmospheric constituents.

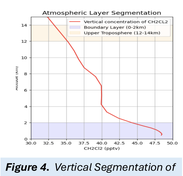


Figure 4. *Vertical Segmentation of Atmospheric Layers*

Data analysis

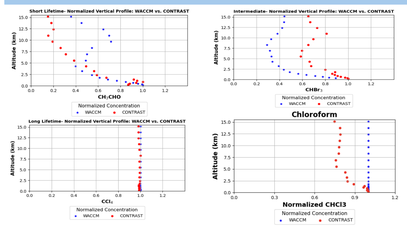
The analysis focuses on the area covered by the CONTRAST campaign, specifically the tropical western Pacific region.
The study examines 26 tracers from WACCM, ranging from very short-lived species like acetaldehyde to extremely long-lived species such as CFC-11, with lifetimes extending up to thousands of years in different atmospheric layers.
Tracer lifetimes (LT) and Transit Time Distributions (TTD) were derived from regional averages across the tropical western Pacific. However, for direct comparisons with airborne observations, WACCM output was interpolated along the CONTRAST flight tracks. This combination of regional and flight-specific analysis enhances the ability to evaluate both broad transport patterns and localized discrepancies along flight paths, offering a more comprehensive assessment of tracer transport by tropical convection.

Vertical Profiles Analysis
Vertical profiles of Very Short-Lived (VSL), Intermediate, and Long-Lived tracers were averaged per kilometer altitude. Data from both WACCM and the CONTRAST data.
The UT:BL ratios were calculated using altitude definitions where the upper troposphere (UT) is defined as the layer between 12-14 km and the boundary layer (BL) is defined as the layer from 0-2 km.

Vertical Profile Shapes of Observed Tracers:
Short-Lived Tracers: Shows high surface concentrations with a rapid decrease at increasing altitudes, underlining its short atmospheric lifetime and limited vertical transport.
Intermediate Tracers: Consistently present in both the boundary layer (BL) and the upper troposphere (UT). Their vertical profiles show significant concentrations at the surface, gradually decreasing but remaining detectable at higher altitudes. This suggests strong convective transport mechanisms. Notably, chloroform shows discrepancies between WACCM simulations and observational data, with WACCM consistently underestimating its variability.
Long-Lived Tracers: Demonstrate uniform distribution throughout the troposphere. Their concentration remains relatively constant from the surface to the upper troposphere, indicating transport by gradual lifting, reflecting their long lifetime and stability in the atmosphere.

Model-Observation Comparison
WACCM simulations and CONTRAST observations show high surface concentrations of tracers decreasing with altitude due to convective transport. However, discrepancies arise for specific tracers like chloroform, where WACCM consistently underrepresents UT concentrations. For VSL species like Acetaldehyde, CONTRAST data show a sharp decline with altitude, while WACCM overestimates UT concentrations, suggesting potential differences in the representation of vertical transport or photochemical loss.

UT:BL vs. TL Relationship



Short-Lived Tracers (<10 days):
Low UT:BL ratios show most short-lived tracers decay in the boundary layer. WACCM predicts higher upward transport than CONTRAST.
Intermediate Tracers (10-300 days):
UT:BL ratios increase with lifetime, reflecting better transport. Differences between WACCM and CONTRAST indicate variability in transport and loss rates.
Long-Lived Tracers (>1000 days):
UT:BL ratios near 1 show even distribution across the troposphere. Strong WACCM-CONTRAST agreement highlights accurate transport representation.

Figure 5. Vertical Profiles Comparing Tracers with Varying Lifetimes: Short-Lived (CH_3CHO), Intermediate ($CHBr_3$), and Long-Lived (CCl_4). The plots display normalized concentrations from WACCM and CONTRAST observations, with WACCM data interpolated along the CONTRAST flight tracks to ensure direct comparison. Concentrations are averaged over 1 km intervals around each altitude level in the WACCM model.

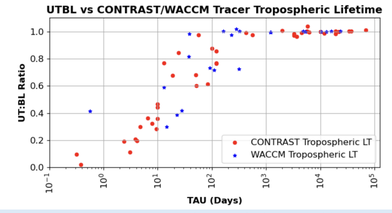
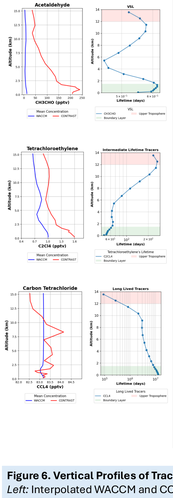


Figure 7. UT:BL Ratio vs. Whole Tropospheric Tracer Lifetime for WACCM and CONTRAST – Lifetime estimates from the paper
Note: Lifetime estimates are taken from Luo and Pan et al., 2018, and Chelpon et al., 2021.

Tracer Groups Based on Lifetime



Very Short-Lived (VSL) Tracers – Acetaldehyde (CH_3CHO) Profile:
Sharp decline above the boundary layer (BL), indicating rapid photochemical removal. Minimal presence in the upper troposphere (UT) suggests limited transport and concentration near emission sources.

Intermediate-Lifetime Tracers Tetrachloroethylene (C_2Cl_4) Profile:
Exhibits a reverse S-shaped vertical profile with enhancements in both the BL and UT. This pattern reflects convective uplift, with partial photochemical loss occurring in the mid-troposphere.

Long-Lived Tracers – Carbon Tetrachloride (CCl_4) Profile:
Displays a uniform vertical distribution, highlighting stability across the troposphere. Minimal photochemical loss suggests extensive global mixing and long-term atmospheric transport.

Table 1. *Tracers categorized by atmospheric lifetime: Very Short-Lived (VSL), Intermediate, and Long-Lived species – Based on WACCM estimates.*

Very Short-Lived (VSL) and Short-Lived Tracers

Species	BL Lifetime (days)	Troposphere Lifetime (days)	UT Lifetime (days)
Acetaldehyde	0.6	0.6	0.5
Benzene	8.5	14.8	34.7
Propane	9.4	22.7	70.6
Bromoform	15.9	13	10.3

Intermediate-Lifetime Tracers

Species	BL Lifetime (days)	Troposphere Lifetime (days)	UT Lifetime (days)
Dibromochloromethane	35.3	38	48.9
Ethane	38.4	92.3	259.7
Bromodichloromethane	38.8	37.4	40
1,2-Dichloroethane	43.9	163	639.1
Tetrachloroethylene	56.7	110.9	205.4
Dibromomethane	86.9	231	726.5
Bromochloromethane	95.3	278.8	944.9
Dichloromethane	104.3	312.8	1108.7
Chloroform	104.5	322.6	1155.6

Long-Lived Tracers

Species	BL Lifetime (days)	Troposphere Lifetime (days)	UT Lifetime (days)
Methyl Chloride	301.5	1200.3	4925.8
Methyl Bromide	350.1	1224.5	4394.7
Methyl Chloroform	1078.6	4863.5	17715.2
HCFC-141b	1856.3	10297.3	45280.3
HCFC-22	2200.5	13020.7	62629.9
HCFC-142b	3184.5	23425.9	120414.4
Carbonyl Sulfide	5291.4	16165.3	51490.2
Halon-1211	7721.2	53238.4	27217
Carbon Tetrachloride	10968652	6055103	62522.3
CFC-113	13695023	79175785	6846996.9
CFC-11	13854190	7675460	113225.3
CFC-12	23750530	13681686	1022775
CFC-114	24440482	14808123	4769889.5

Figure 6. Vertical Profiles of Tracer Lifetimes and Concentrations by Altitude.
Left: Interpolated WACCM and CONTRAST concentrations, averaged over 1 km along the flight track.
Right: Tracer lifetimes by altitude, showing vertical variations across tracer groups.

Sensitivity Analysis

Transport Time Distributions Analysis
TTDs measure the time it takes for tracers to move from the boundary layer (0-2 km) to the upper troposphere (12-14 km), providing insights into the efficiency of convective uplift and tracer transport variability.
TTDs were calculated using three approaches:
BL Lifetimes – Captures rapid convective transport from the lower troposphere.
UT Lifetimes – Reflects the transport of tracers reaching the upper troposphere.
Full Tropospheric Lifetimes – Provides an integrated view of transport across the entire troposphere.
Tropospheric lifetimes offer the best overall representation for TTD, while BL and UT-derived TTDs complement the analysis by highlighting specific transport processes and enabling model-observation comparisons.

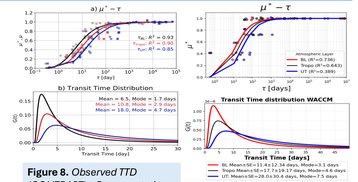


Figure 8. *Observed TTD (CONTRAST) – Representation of Transport Dynamics*
Note: This figure is adapted from Chelpon et al., 2021.

Figure 9. *WACCM TTD - Model-Calculated Transport Patterns.*

Transport Time Distributions (TTDs) varies by tracer's lifetime.
Short-lived tracers show sharp TTD peaks, indicating rapid vertical transport, while longer-lived tracers have broader distributions.
WACCM captures general transport patterns but slightly differs from observations. The model captures rapid convective transport in the boundary layer but shows more scattered transport in the upper troposphere.
This suggests potential differences in the model's representation of high-altitude convective processes, warranting further investigation.

Impact of VOC Lifetime Thresholds on TTD Variability

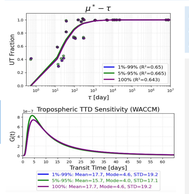


Figure 10. *Outlier filtering (5%-95%) reduces noise and narrows TTDs, preserving overall structure and mean.*

Filtering outliers (5%-95%) reduces noise and narrows TTDs, primarily affecting the spread without changing the mean or mode.
Minimal differences indicate large-scale variability dominates, preserving the overall TTD structure.

Comparison of TTDs with Different Initial Source Regions

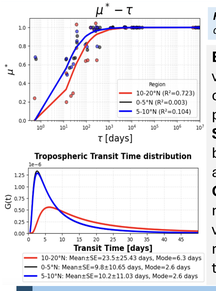


Figure 11. *Sensitivity of TTDs to regional differences*

Equatorial regions show faster vertical transport due to strong convection, resulting in sharp TTD peaks and shorter transit times.
Subtropical zones have slower, broader TTDs, reflecting weaker ascent and more stable transport.
Convection drives rapid transport near the equator, large-scale variability influences subtropical regions, leading to slower overall transport.

Conclusion and discussion

Tracer Behavior and Transport Dynamics:
Short-Lived Tracers: High near-surface concentrations that decrease sharply with altitude illustrate rapid boundary layer decay.
Intermediate Tracers: Significant concentrations from the surface to the upper troposphere indicate of effective convective uplift.
Long-Lived Tracers: Stable concentrations across altitudes, showing the effectiveness of steady vertical transport.

Model-Observation Integration:
The goal of further analysis is to evaluate and quantify the model's representation of atmospheric processes, particularly convective uplift and tracer distribution. By integrating airborne observations and model outputs, this study aims to improve understanding of transport by tropical convection.

Continuous Model Improvement:
Ongoing efforts focus on comparing WACCM simulations with airborne data to assess discrepancies and refine the model's performance. This process will help identify variability and factors contributing to differences in tracer profiles, particularly for short-lived species.

Sensitivity Analysis:
Adjustments to VOC lifetimes and source region treatments revealed significant impacts on modeled TTDs, emphasizing the model's sensitivity to these parameters. Further analysis will refine these inputs to enhance simulation accuracy.

Acknowledgements and references

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