

Physics-Informed Machine Learning for Global Stratospheric Composition (1979–2026)

Sandip S. Dhomse^{1,2} & Martyn P. Chipperfield^{1,2}

¹ School of Earth, Environment and Sustainability, University of Leeds, Leeds, UK ² NCEO, University of Leeds, UK s.s.dhomse@leeds.ac.uk

1. Scientific Challenge & Objective

Satellite observations provide high-quality stratospheric profiles but remain spatially and temporally sparse, while chemistry transport models provide complete 3D fields but contain systematic uncertainties associated with transport, meteorology, and chemical parameterisations (Dhomse et al., 2021, 2023, 2026).

Objective: combine satellite observations, TOMCAT, and machine learning to construct continuous, observation-constrained global stratospheric composition fields.

- **TCOM v2.0 (released):** XGBoost correction of TOMCAT using ACE-FTS v5.3 observations, producing daily 3D fields for ten stratospheric species.
- **PINN-TCOM (in development):** physics-informed temporal learning that embeds transport and chemistry constraints to support reconstruction across data-sparse periods extending back to 1979.
- **Key Applications:** Serves as a high-fidelity benchmark for **chemical model evaluation**, a prior/constraint for **satellite retrieval algorithms**, and a foundational dataset for training atmospheric **Digital Twins**.

Data access: TCOM v2.0 datasets for O₃, H₂O, CH₄, N₂O, CFC-11, CFC-12, HF, HCl, HNO₃, and COF₂ are publicly archived on Zenodo. Project info: <https://tomcat.leeds.ac.uk/tomcat/tcom/>

2. Observations & Model Inputs

Integration of multi-decadal observational satellite data and chemistry transport modelling:

- **ACE-FTS Satellite Data (v5.3):** High-resolution infrared solar occultation profiles (~30 profiles/day, 1.5–4 km vertical resolution, 2004–2025) (Bernath et al., 2005; Boone et al., 2023).
- **Legacy Satellite Records:** Historical solar occultation profiles from SAGE I (1979–1981), SAGE II (1984–2005), and HALOE (1991–2005) to extend series back to 1979.
- **TOMCAT 3D CTM:** Offline CTM forced by ERA5 reanalyses (2.8° × 2.8° resolution, 32/60 vertical levels up to ~60 km) (Chipperfield, 2006; Hersbach et al., 2020).
- **SPARC Data Initiative:** Multi-satellite compilations (ACE, MIPAS, HIRDLS, SWOOSH) for independent validation (Hegglin et al., 2021).

3. Why XGBoost? (TCOM v2.0)

Rigorous evaluation across seven regression algorithms (OLS, Lasso, Ridge, ElasticNet, Random Forest, AdaBoost, XGBoost) identified XGBoost as the optimum model due to its gradient boosting architecture and built-in L1/L2 regularisation (Dhomse & Chipperfield, 2026).

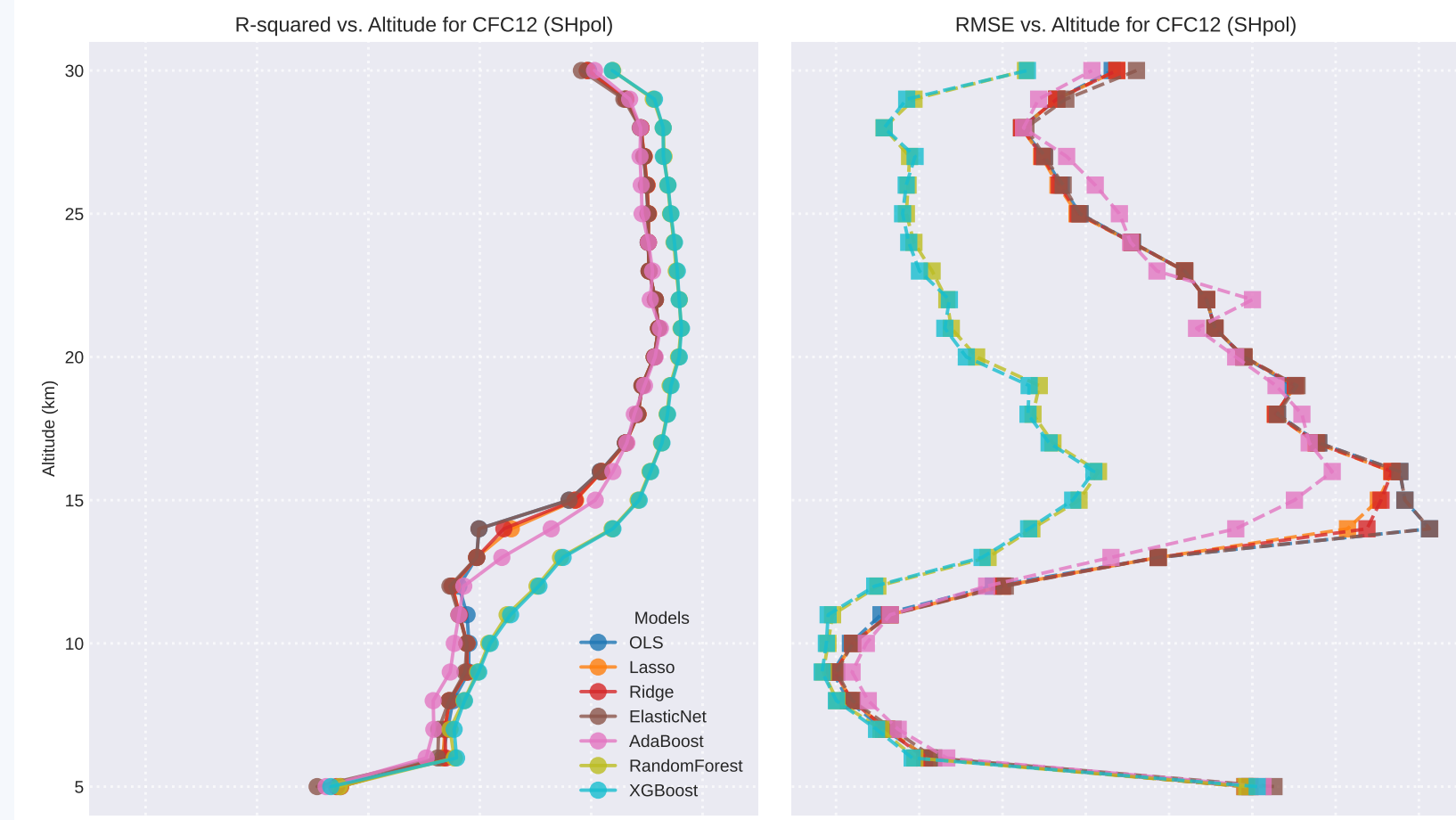


Figure 1: Algorithm benchmark for CFC-12 in SH polar region (60°S–90°S). Metrics show R^2 (left) and RMSE (right) from 5 to 30 km. Linear models show poor fit ($R^2 < 0.50$) below 15 km, whereas XGBoost achieves $R^2 > 0.90$ and minimum RMSE across all altitude layers.

4. XGBoost Skill Across Latitude & Altitude

XGBoost performance metrics for CFC-12 across all 5 latitude bins demonstrate high predictive skill throughout the stratosphere: **Model Performance Metrics for CFC12**

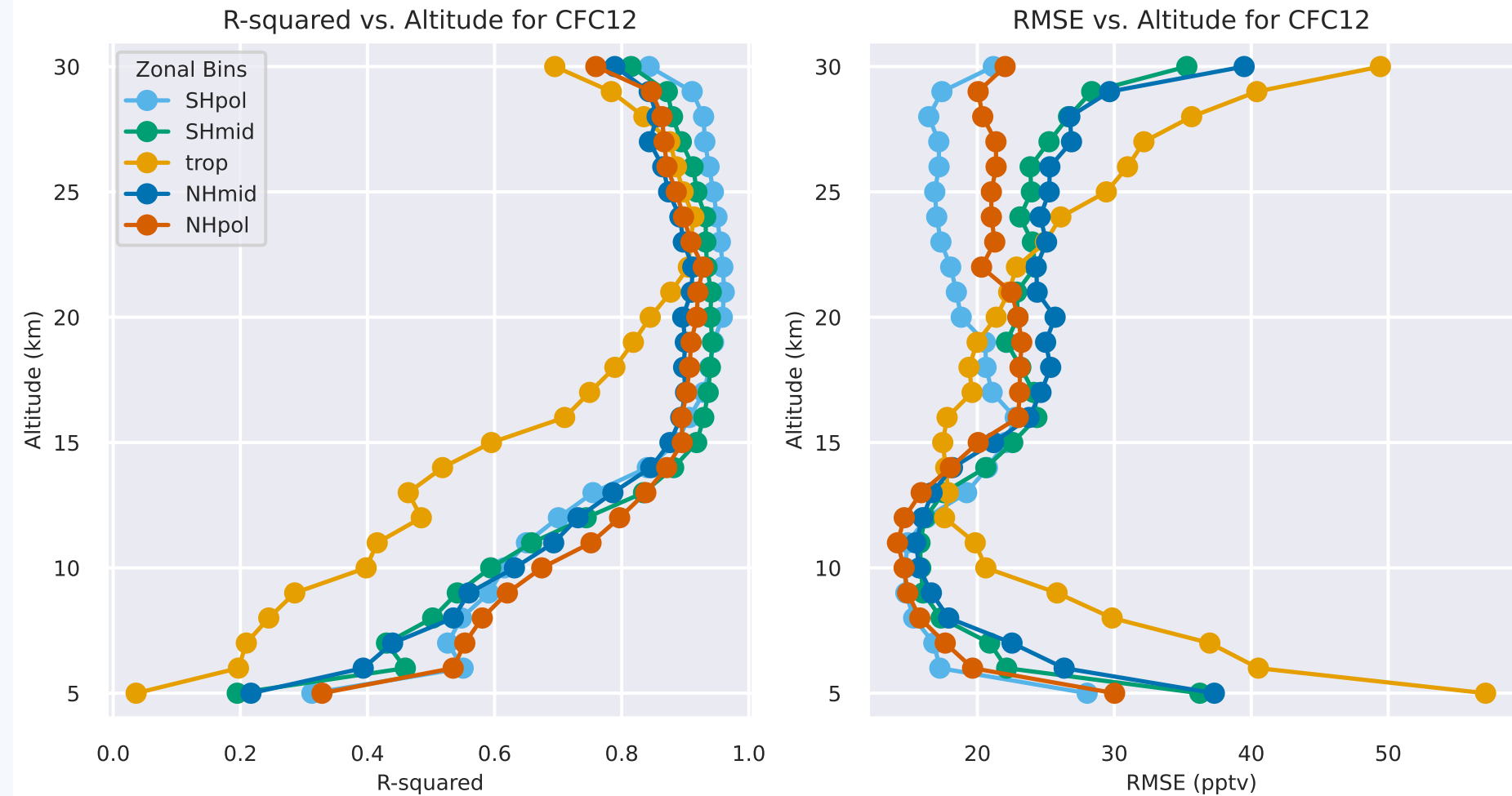


Figure 2: Vertical profiles of R^2 (left) and RMSE (pptv, right) for XGBoost predicting CFC-12. $R^2 > 0.85$ – 0.95 in mid-latitude and polar zones; lower tropical R^2 near 15–18 km reflects TTL convective mixing and QBO dynamics. Lower tropical skill is due to fewer ACE-FTS measurements from satellite orbit design.

5. TCOM Corrects TOMCAT Bias

Model Analysis for CFC12 in SHpol at 14 km

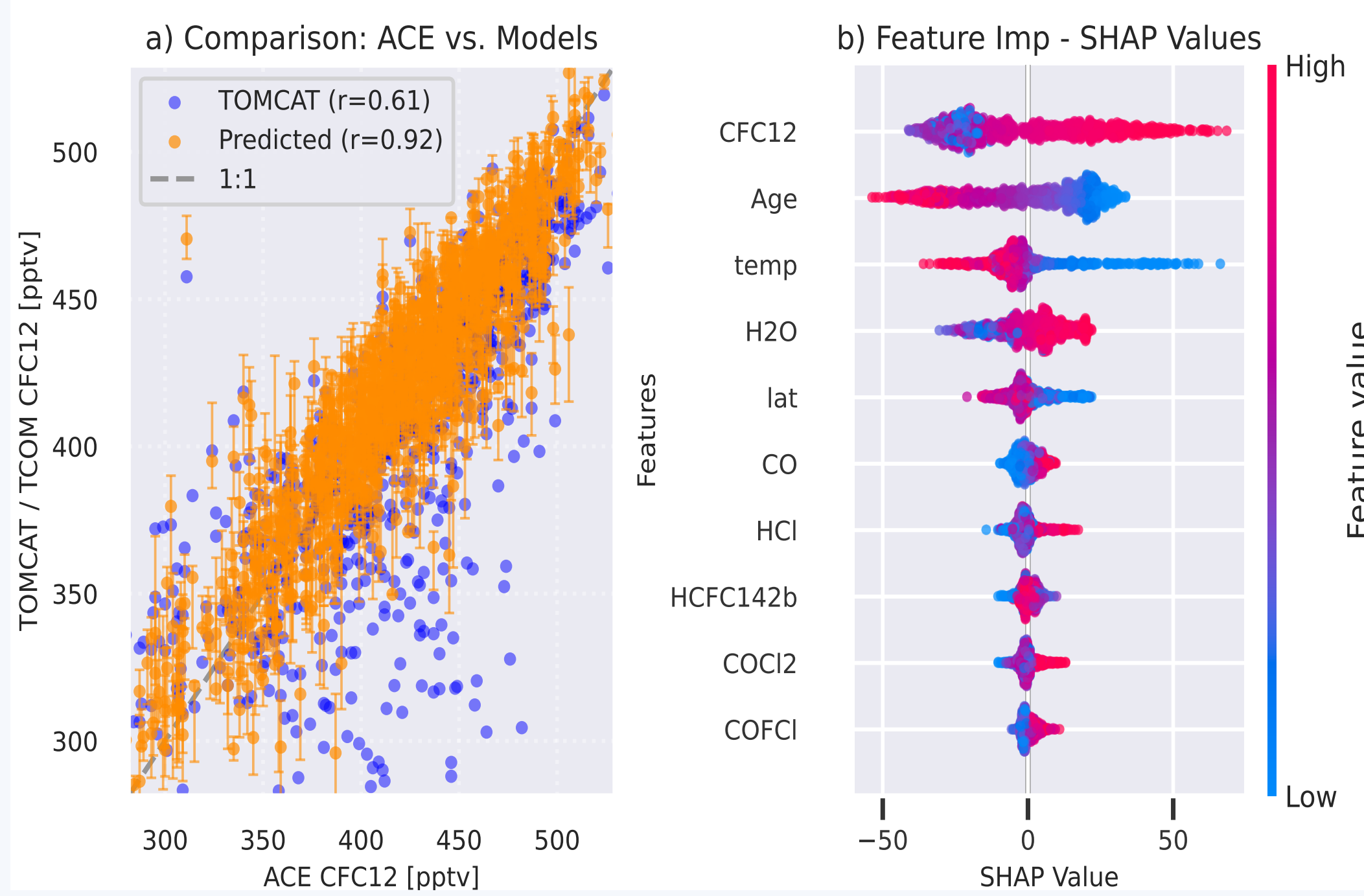


Figure 3: (a) Scatterplot comparing ACE-FTS vs. TOMCAT CTM (blue) and TCOM XGBoost (orange) for CFC-12 at 14 km (SHpol). Bias correction raises Pearson r from 0.61 to 0.92. (b) SHAP ranking shows TOMCAT CFC-12 as the most important feature, while Age-of-Air (AoA) and H₂O act as transport correctors.

6. Independent Multi-Satellite Evaluation

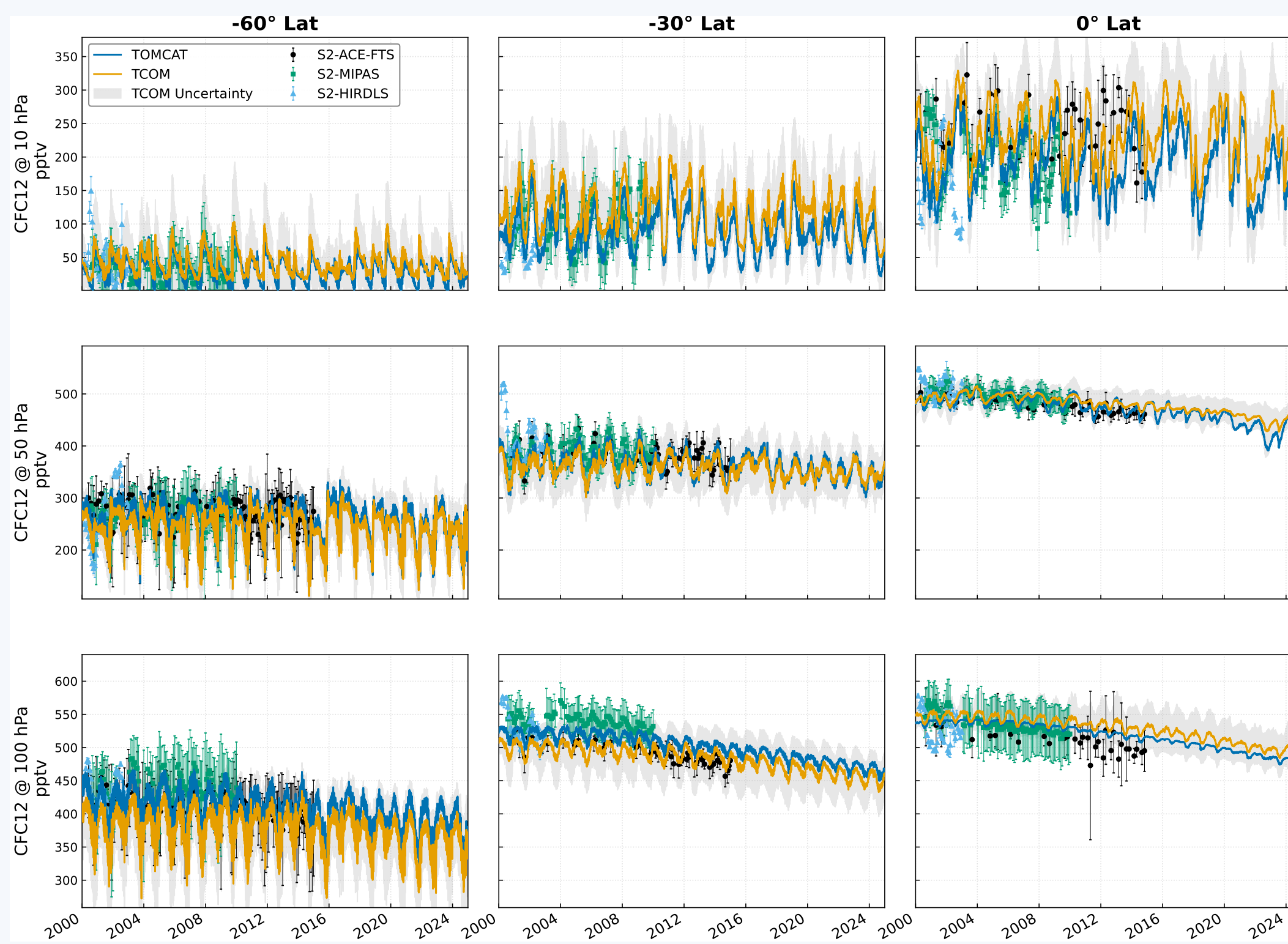


Figure 4: Temporal comparison of daily-mean CFC-12 VMRs (pptv) from 2000 to 2004 across 3 latitudes (60°S, 30°S, 0°) at 10 hPa, 50 hPa, and 100 hPa vs satellite records (ACE-FTS, MIPAS, HIRDLS) (Hegglin et al., 2021). TCOM v2.0 resolves raw CTM negative biases, matching SPARC v2 observations within $\pm 1\sigma$.

7. From Statistical Correction to Physics-Informed ML

We are extending TCOM using a Physics-Informed Neural Network with a Temporal Fusion Transformer (TFT) backbone to constrain learning in data-sparse periods:

- **Physics-informed objective:**

$$\mathcal{L}_{\text{total}} = \mathcal{L}_{\text{data}} + \lambda_1 \mathcal{L}_{\text{transport}} + \lambda_2 \mathcal{L}_{\text{chemistry}}$$

- **3D transport constraint ($\mathcal{L}_{\text{transport}}$):**

$$\mathcal{L}_{\text{transport}} = \sum_k \left\| \frac{\partial \chi_k}{\partial t} + \mathbf{u} \cdot \nabla \chi_k - \nabla \cdot (\mathbf{K} \nabla \chi_k) - P_k(\chi) + L_k(\chi) \right\|^2$$

- **Photochemical context:** Solar Zenith Angle (SZA), dynamical tracers, and total halogens (Cl, Br, F) provide chemical constraints.
- **Observational constraints:** legacy occultation records (H₂O, O₃) constrain periods with sparse modern observations.

8. PINN-TCOM Reproduces Observed Vertical Structure

PINN-TCOM retains physically realistic ozone profile structure while correcting systematic departures from ACE-FTS observations:

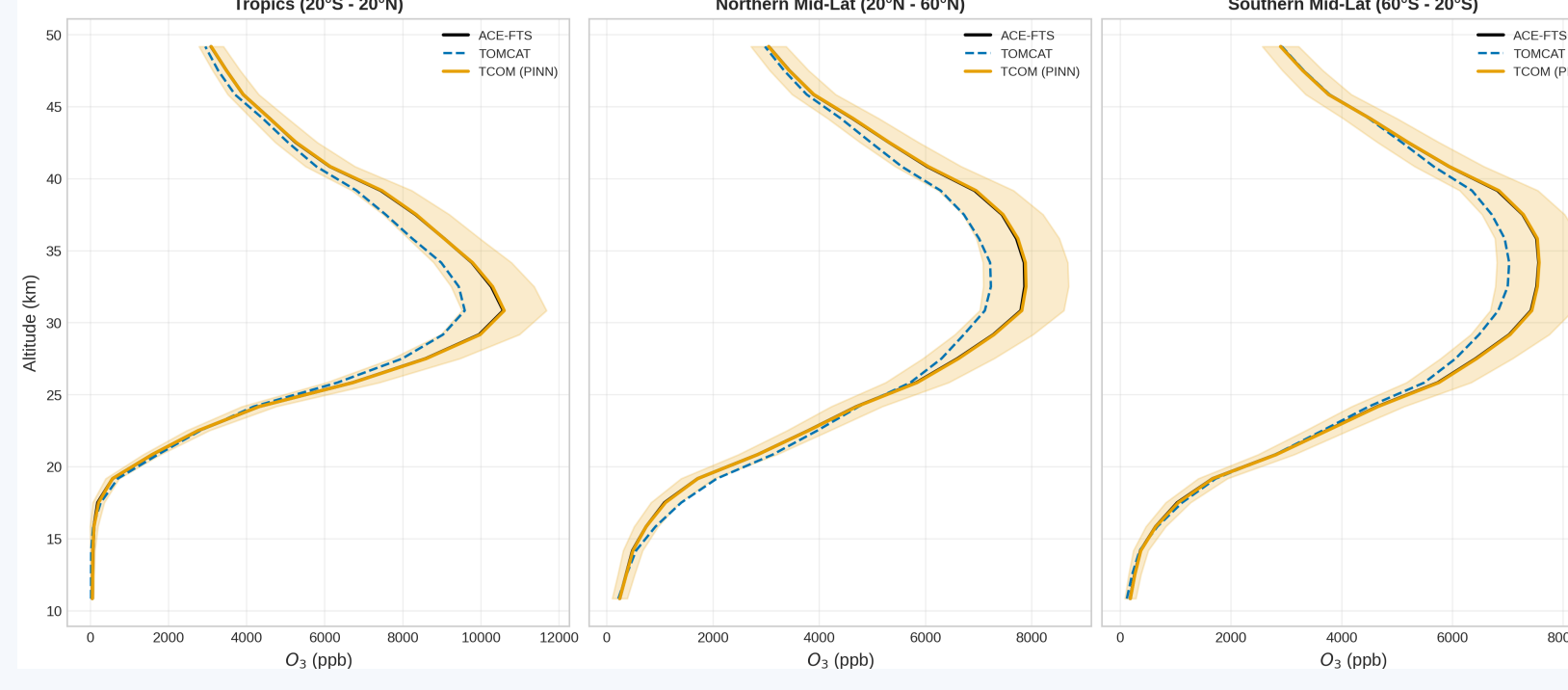


Figure 5: Vertical profiles of stratospheric ozone mixing ratios comparing raw TOMCAT output, ACE-FTS observations, and multi-seed PINN-TFT predictions. Shaded envelopes denote combined epistemic uncertainty (inter-seed variance) and aleatoric noise tracking profiles from 20 km to 45 km.

9. PINN-TCOM Improves Agreement Across Domains

Across latitude–altitude domains, PINN-TCOM shows improved agreement with ACE-FTS relative to the baseline TOMCAT simulation:

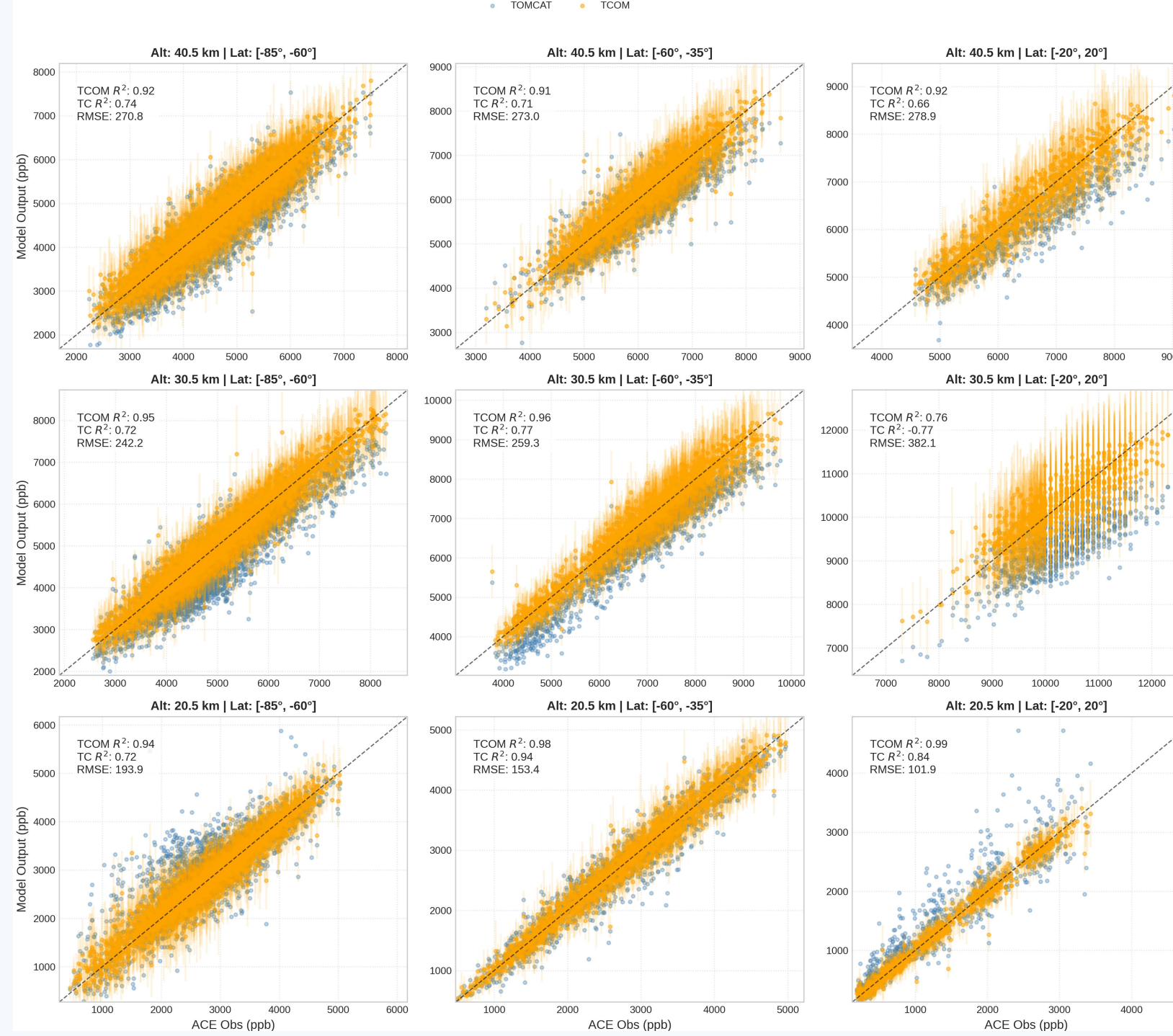


Figure 6: Baseline TOMCAT vs. PINN-corrected predictions against ACE-FTS across latitudes and stratospheric altitudes (40.5 km, 30.5 km, 20.5 km). PINN predictions cluster tightly along the 1 : 1 line, with tropical lower stratospheric RMSE dropping to 101.9 ppb.

10. Global Spatial Performance

Latitude–altitude diagnostics identify where the reconstruction improves upon TOMCAT and where residual errors remain:

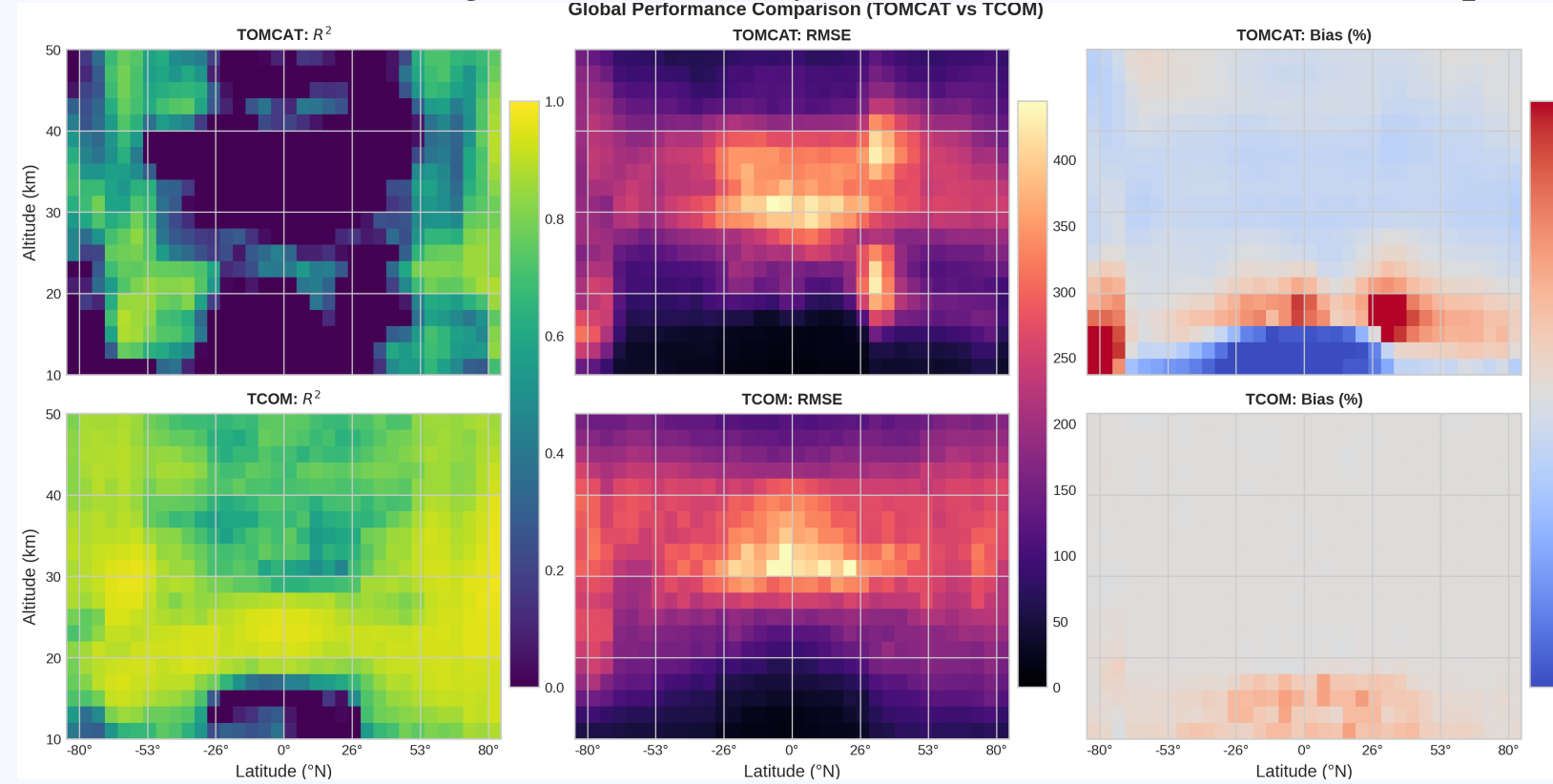


Figure 7: Latitude–altitude performance maps comparing TOMCAT and the corrected reconstruction using R^2 , RMSE, and bias. The diagnostic highlights where ML correction provides the largest improvement and where residual model error remains.

11. Preliminary Tropical Ozone Reconstruction

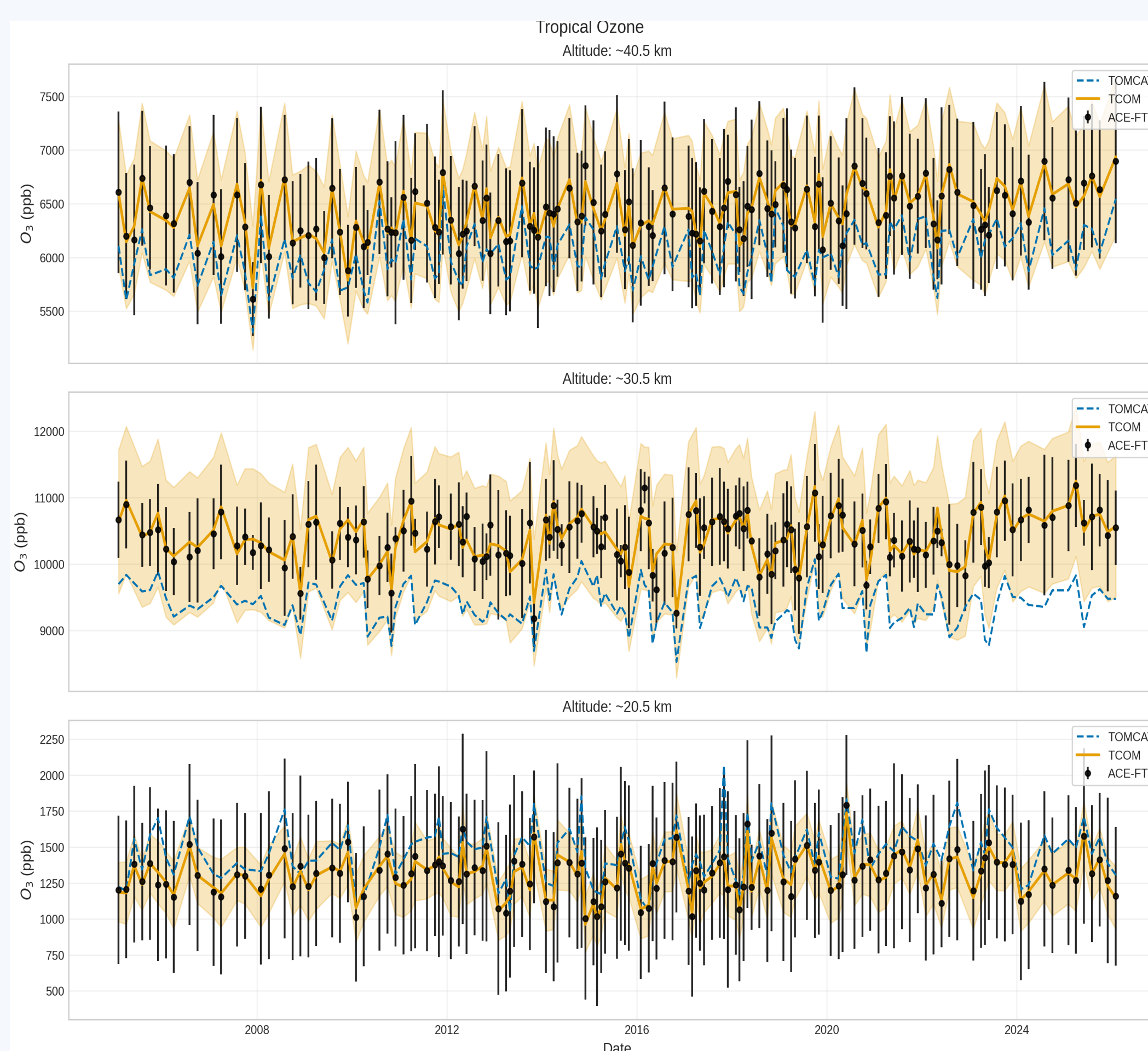


Figure 8: Preliminary tropical ozone reconstruction spanning the ACE-FTS observation period (~2004–2024) across three stratospheric altitudes (40.5 km, 30.5 km, and 20.5 km) within 20°S–20°N. Successfully captures seasonal variability and multi-annual oscillations under physics-informed constraints.

12. Take-Home Messages

- **10 species, global daily 3D fields:** TCOM v2.0 provides observation-constrained reconstructions of O₃, H₂O, CH₄, N₂O, CFC-11, CFC-12, HF, HCl, HNO₃, and COF₂.
- **Effective TOMCAT correction:** XGBoost substantially reduces systematic model–observation differences while retaining physically meaningful CTM information (acts as a “Transport-Corrector”).
- **Physics-informed extension:** PINN-TCOM combines observations with transport/chemistry constraints and explicitly represents predictive uncertainty.
- **Long-term objective:** integrate legacy occultation observations with TOMCAT to support a consistent stratospheric record back to 1979.
- **Broad Utility & Digital Twins:** Reconstructed fields serve as benchmarks for chemical CTM evaluation, prior/constraints for satellite retrieval algorithms, and target datasets for training atmospheric digital twins.

13. Key References

1. Dhomse, S. S. & Chipperfield, M. P. (2026). *Earth Syst. Sci. Data*, 18, 6171–6189.
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