

Supplementary Figures - A comprehensive evaluation of the use of Lagrangian particle dispersion models for inverse modeling of greenhouse gas emissions

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Supplementary Figures S1-S36 illustrate the three investigated baseline methods (REBS method, Stohl's method, and GDB method) at all considered *in situ* measurement sites. The GDB method is illustrated for all tested backward simulation periods, including a case without any backward simulation (0 days). In this extreme case the baseline is obtained directly from the value of the global mixing ratio field simulated with FLEXPART CTM in the spatio-temporal grid cell of the respective observation. REBS and Stohl's method are illustrated for backward simulation periods of 1, 10, and 50 days. Baseline mixing ratios are plotted together with respective observations and *a priori* mixing ratios (sum of the baseline and direct emission contributions).

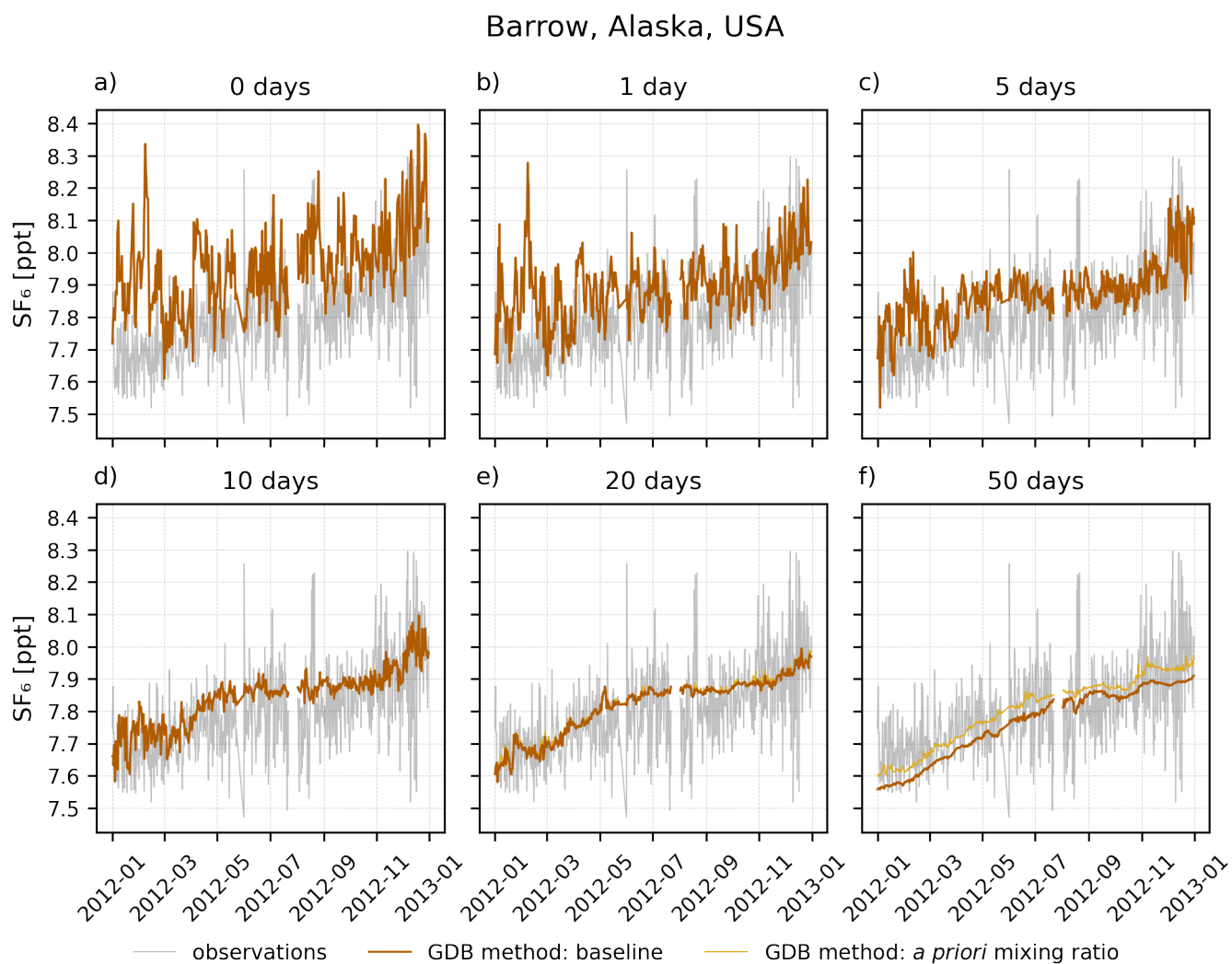


Figure S1: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Barrow observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

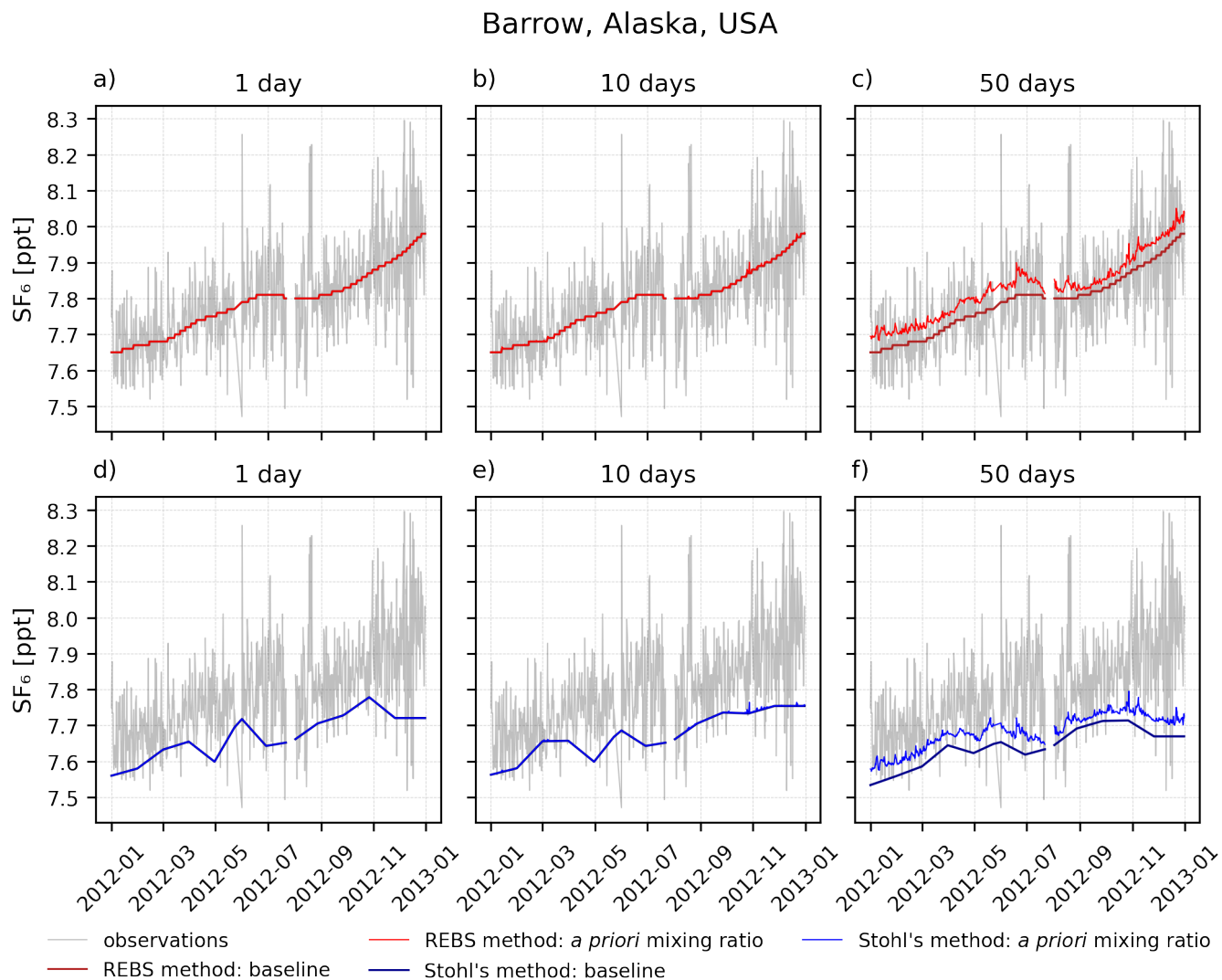


Figure S2: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Barrow observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

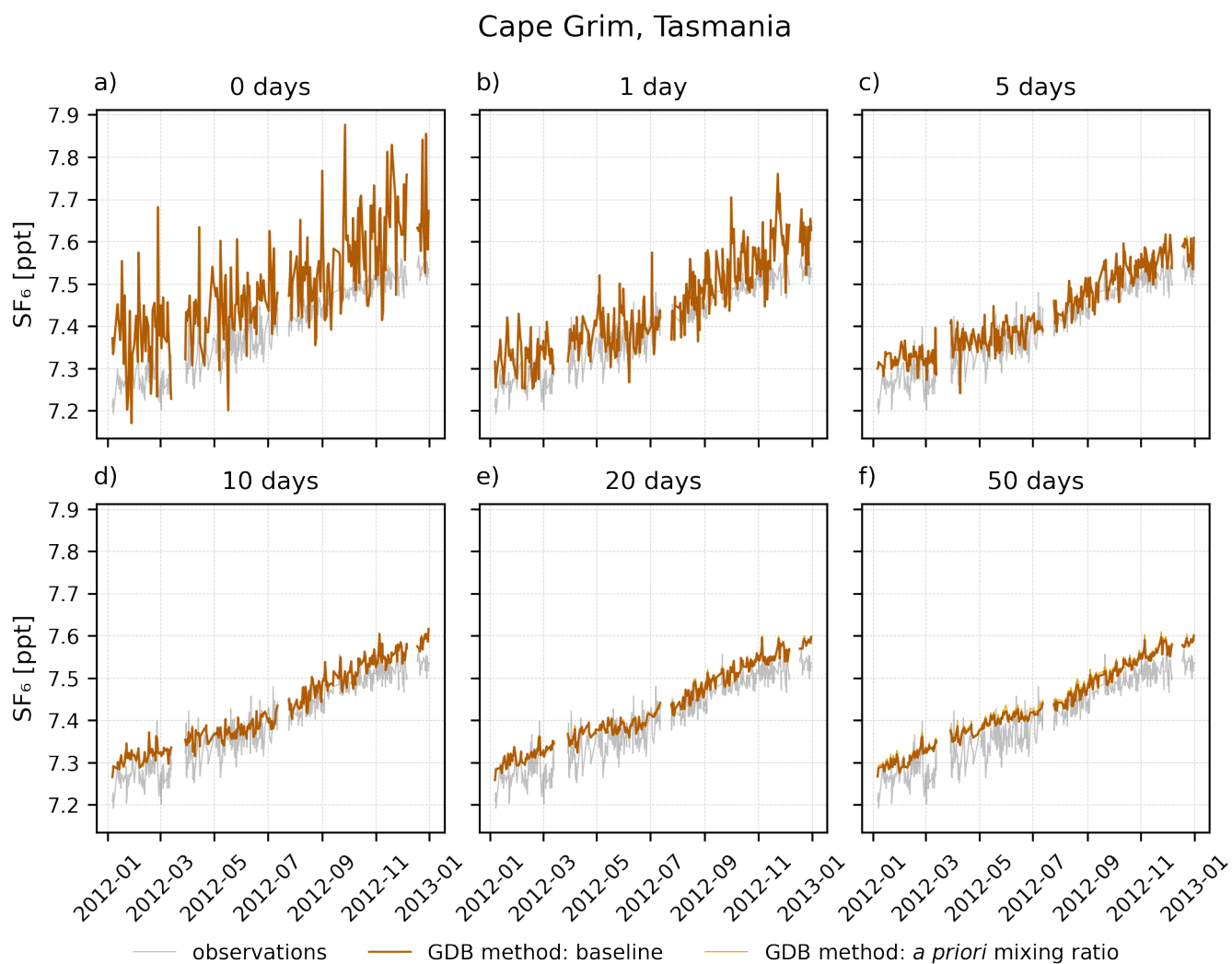


Figure S3: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Cape Grim observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

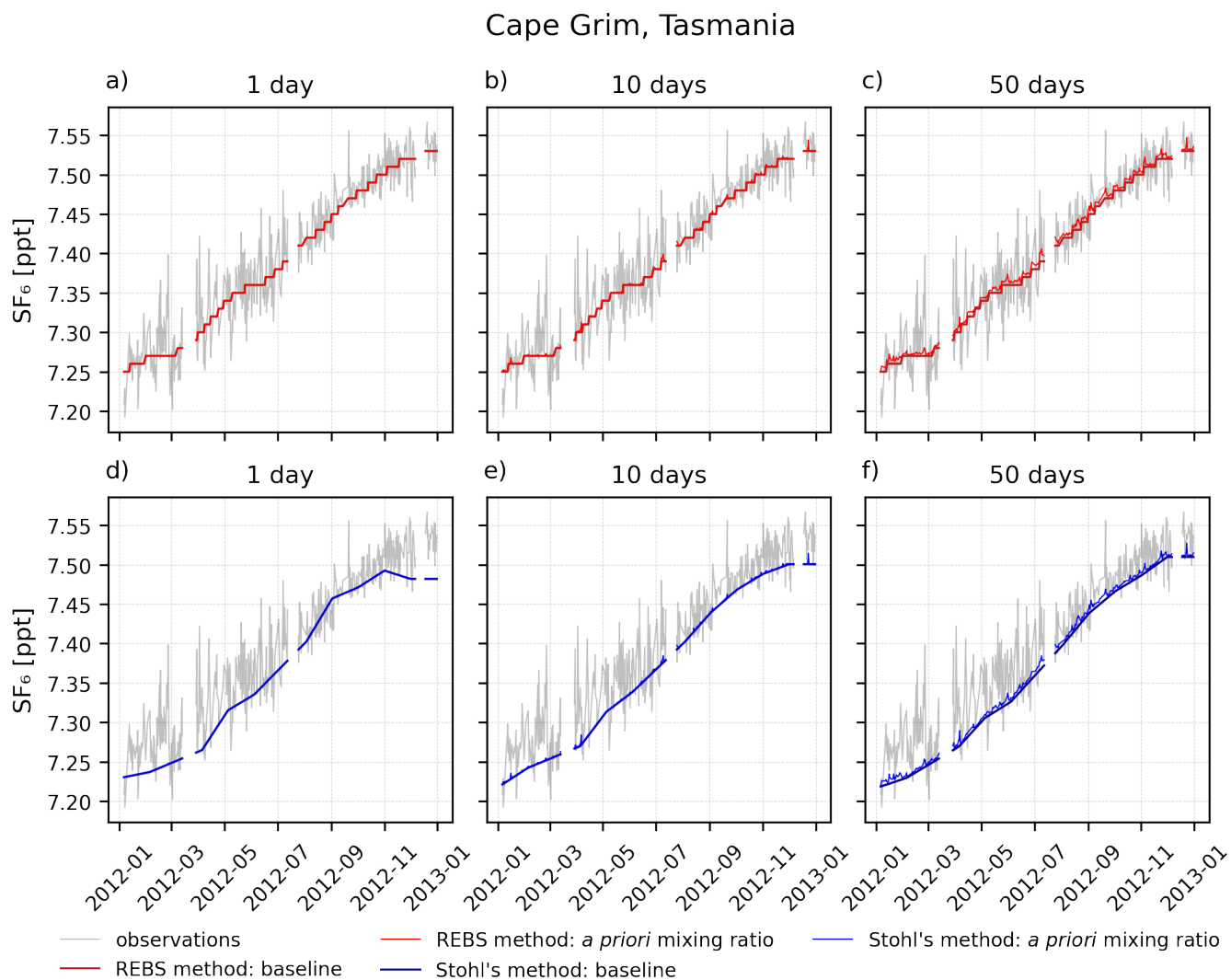


Figure S4: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Cape Grim observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

Cape Matatula, American Samoa

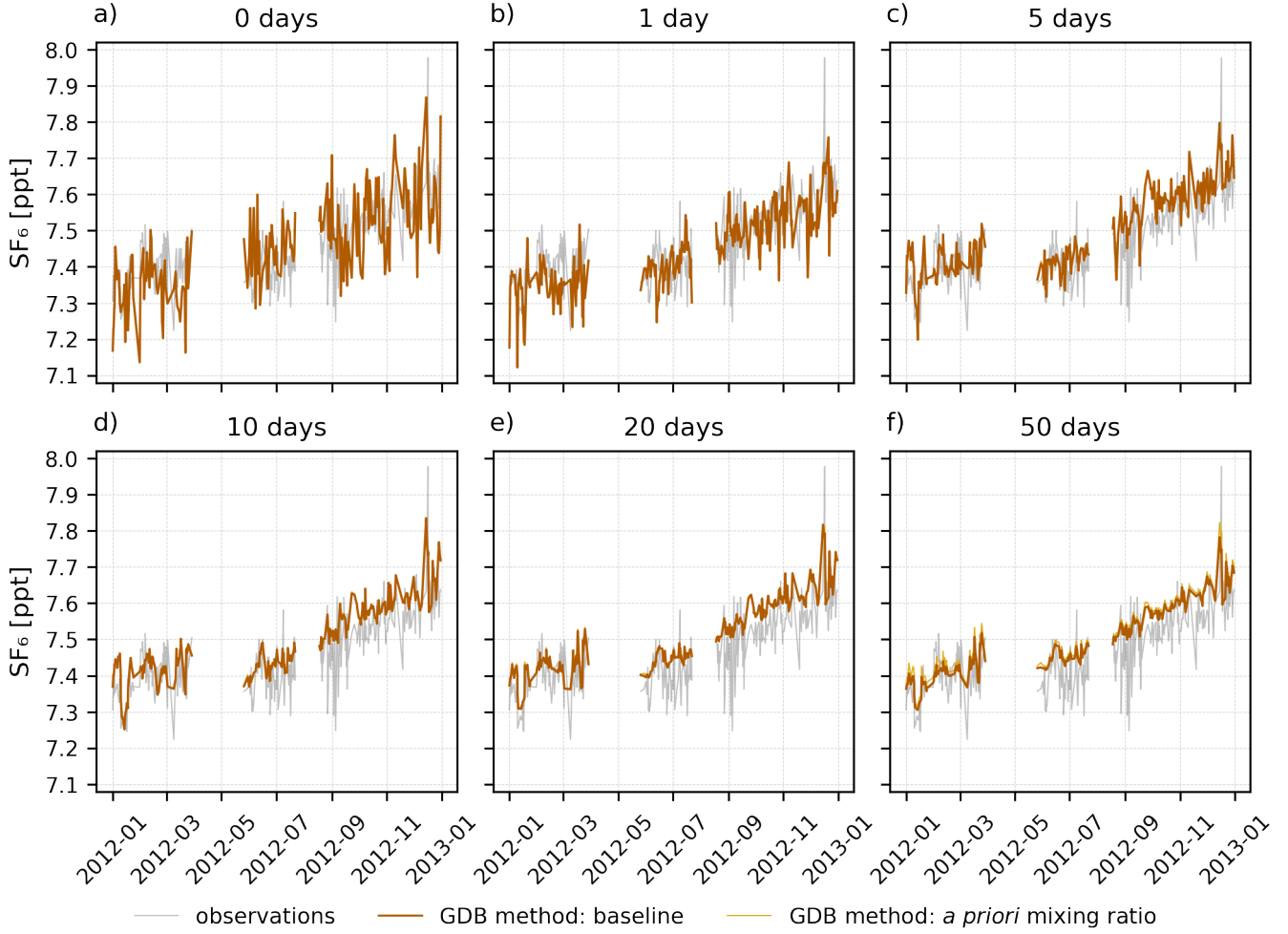


Figure S5: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Cape Matatula observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

Cape Matatula, American Samoa

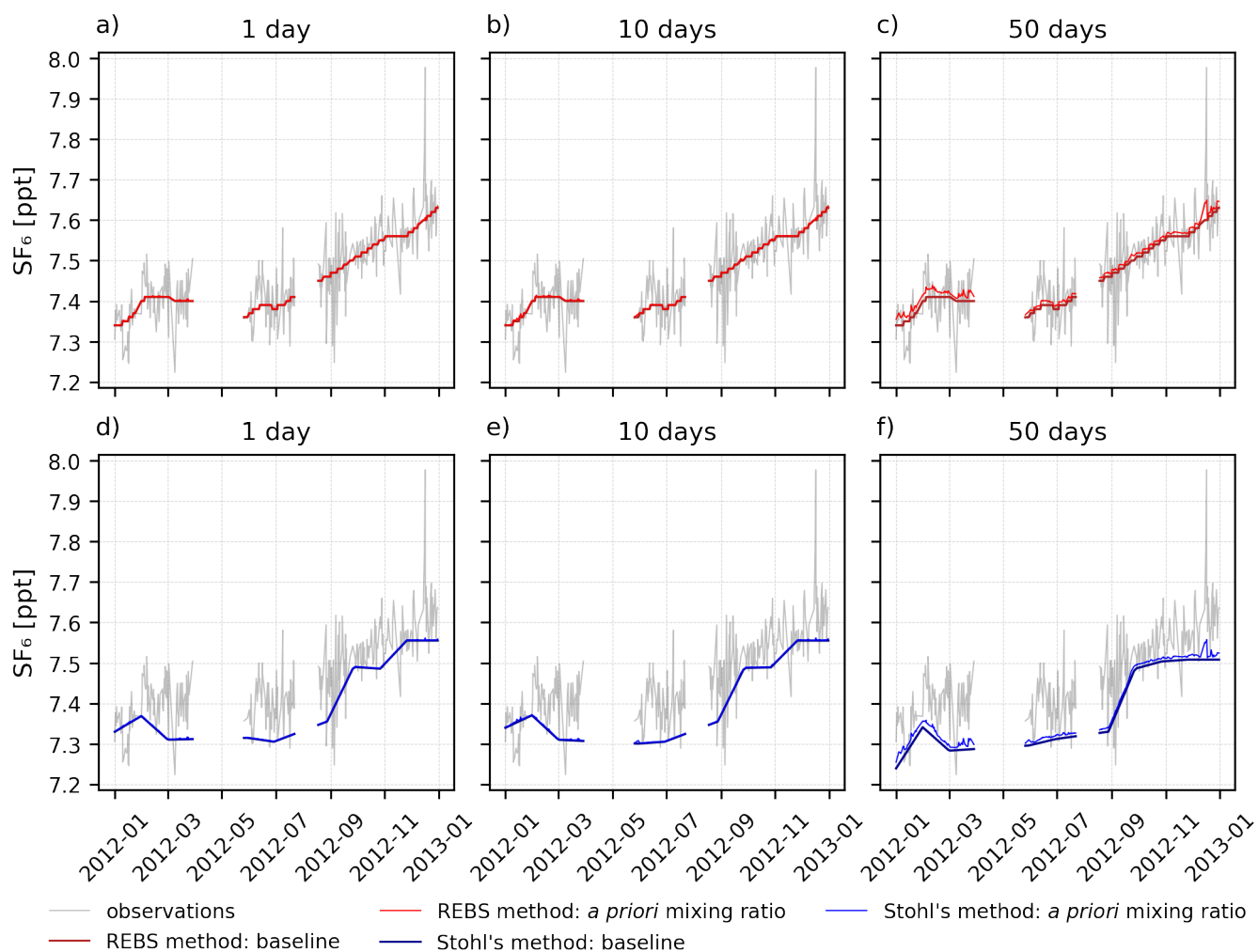


Figure S6: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Cape Matatula observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

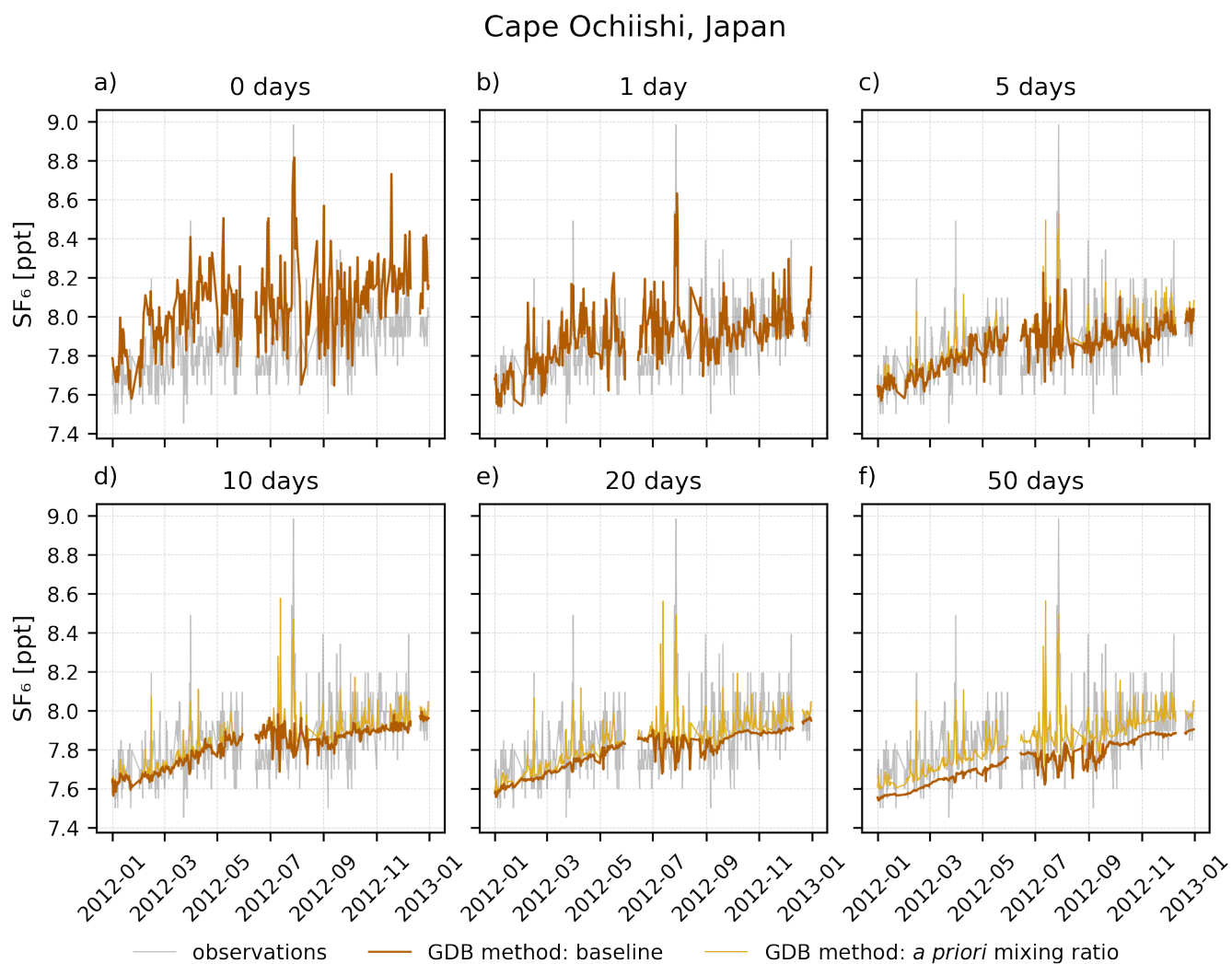


Figure S7: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Cape Ochiishi observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

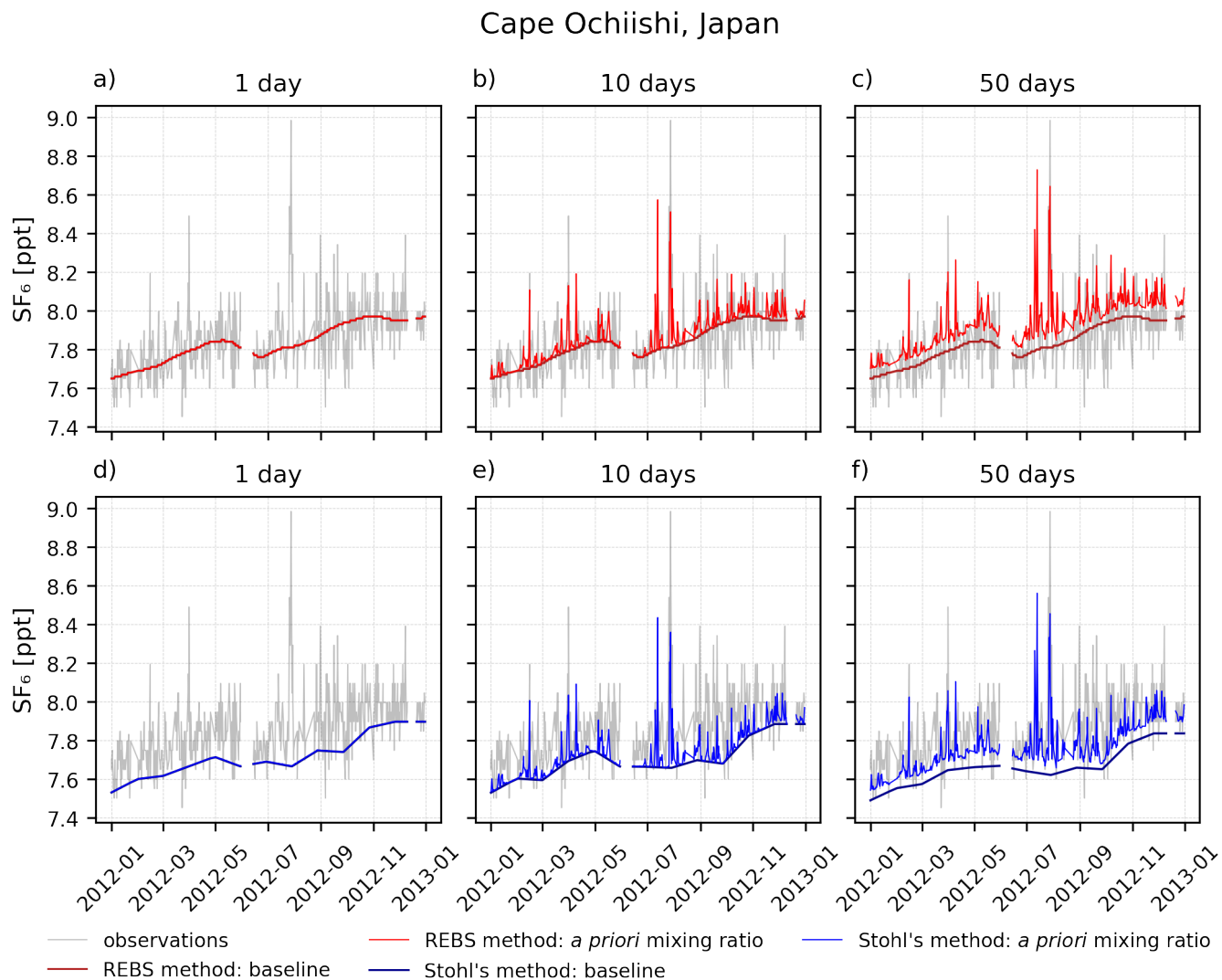


Figure S8: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Cape Ochiishi observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

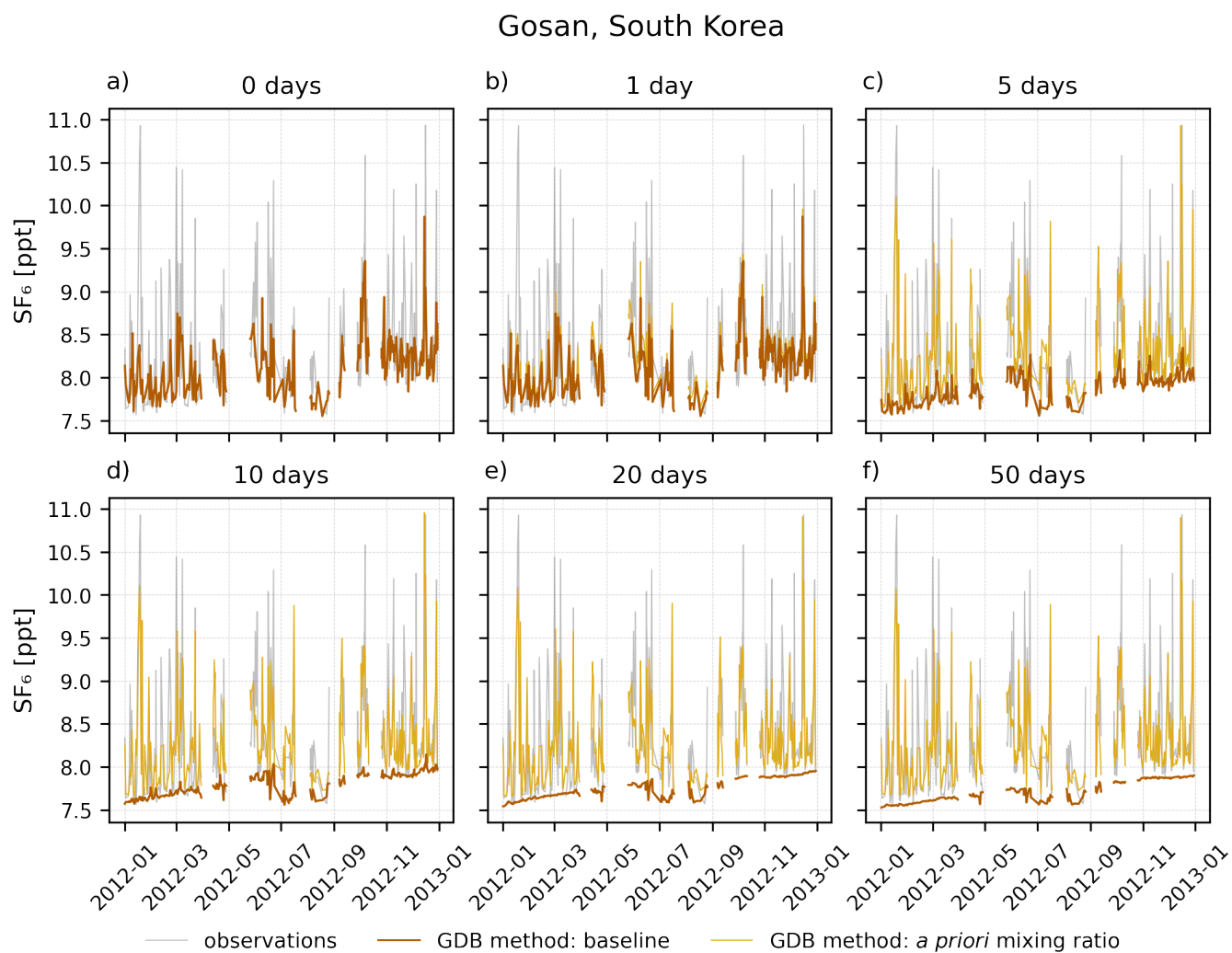


Figure S9: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Gosan observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

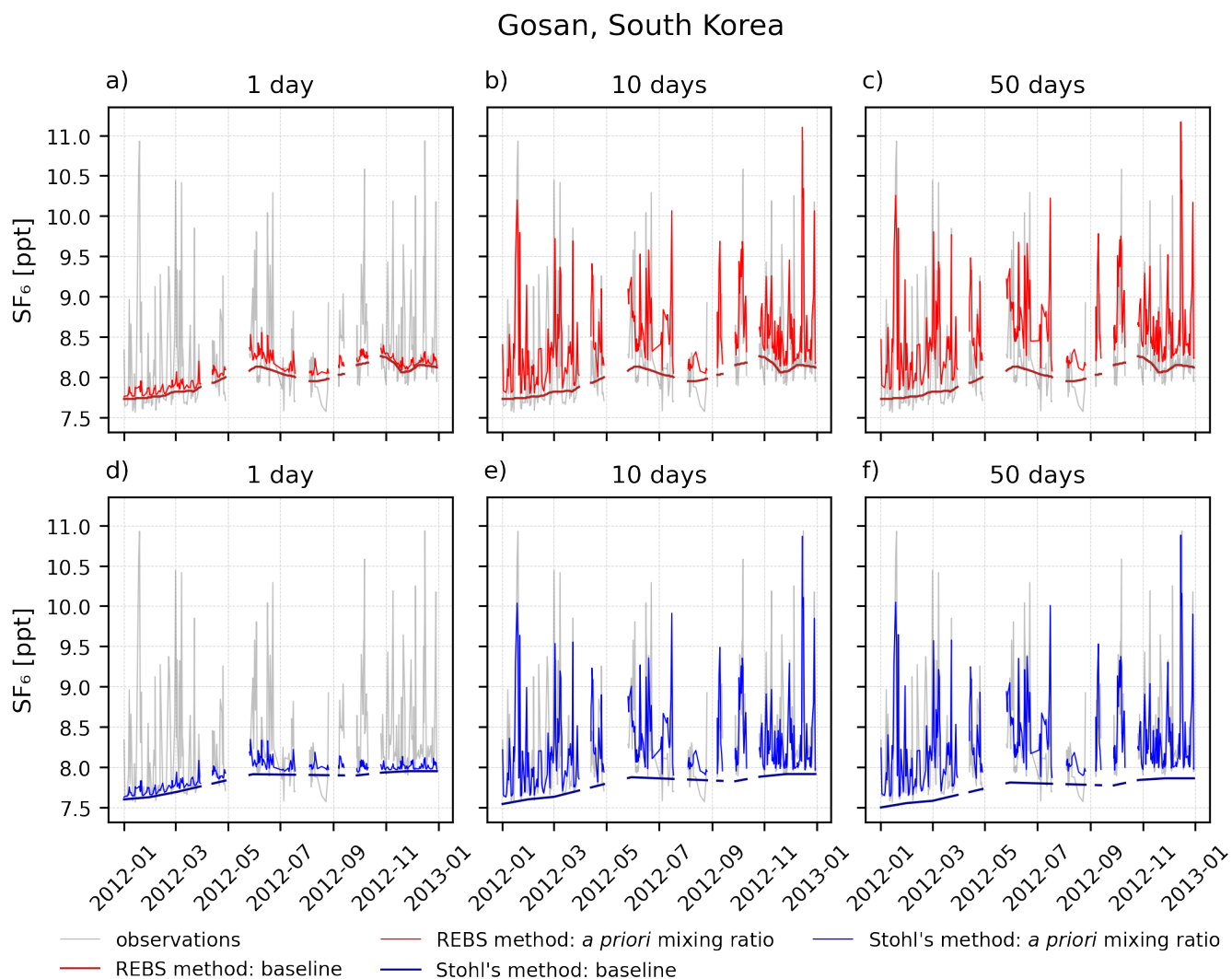


Figure S10: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Gosan observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

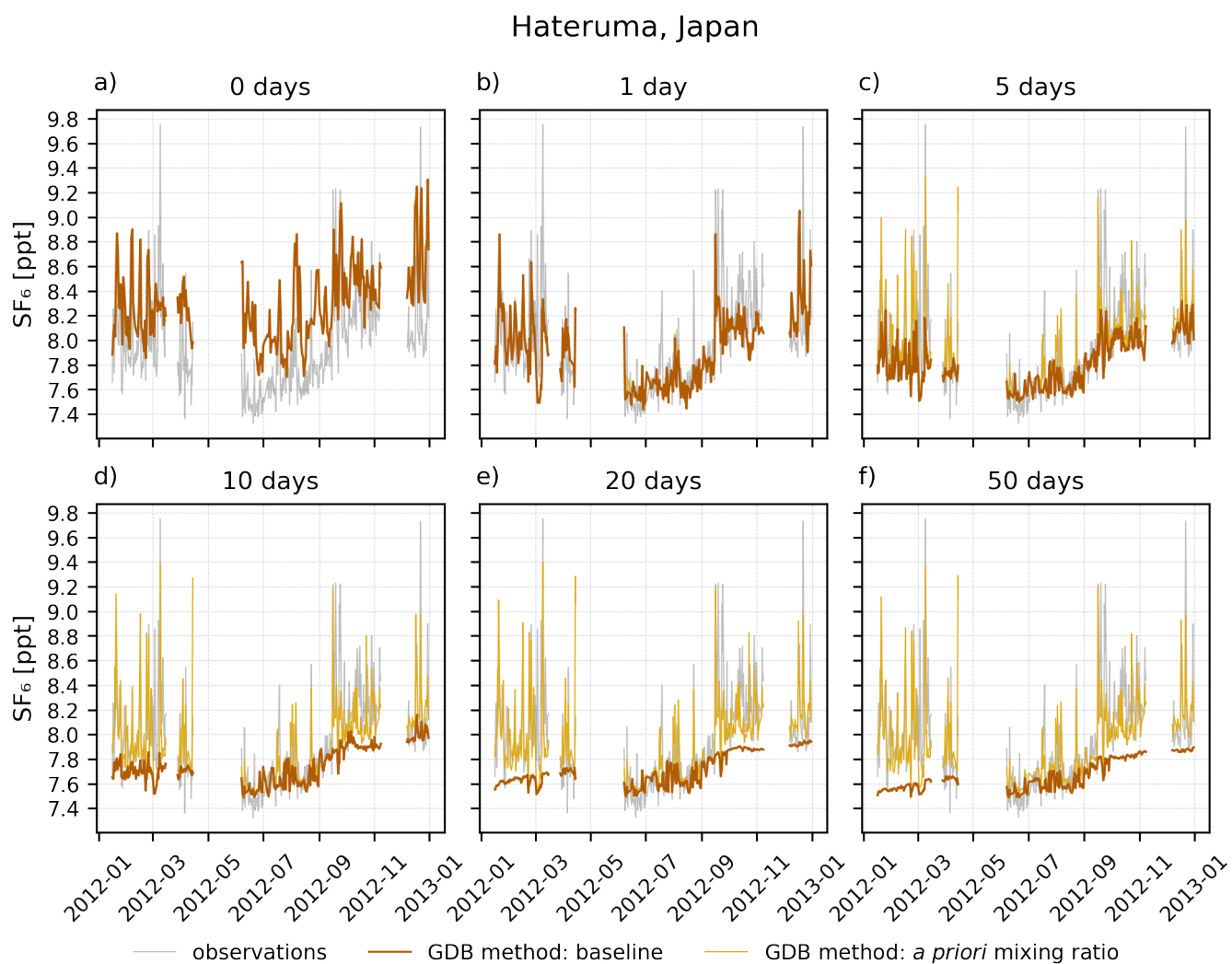


Figure S11: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Hateruma observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

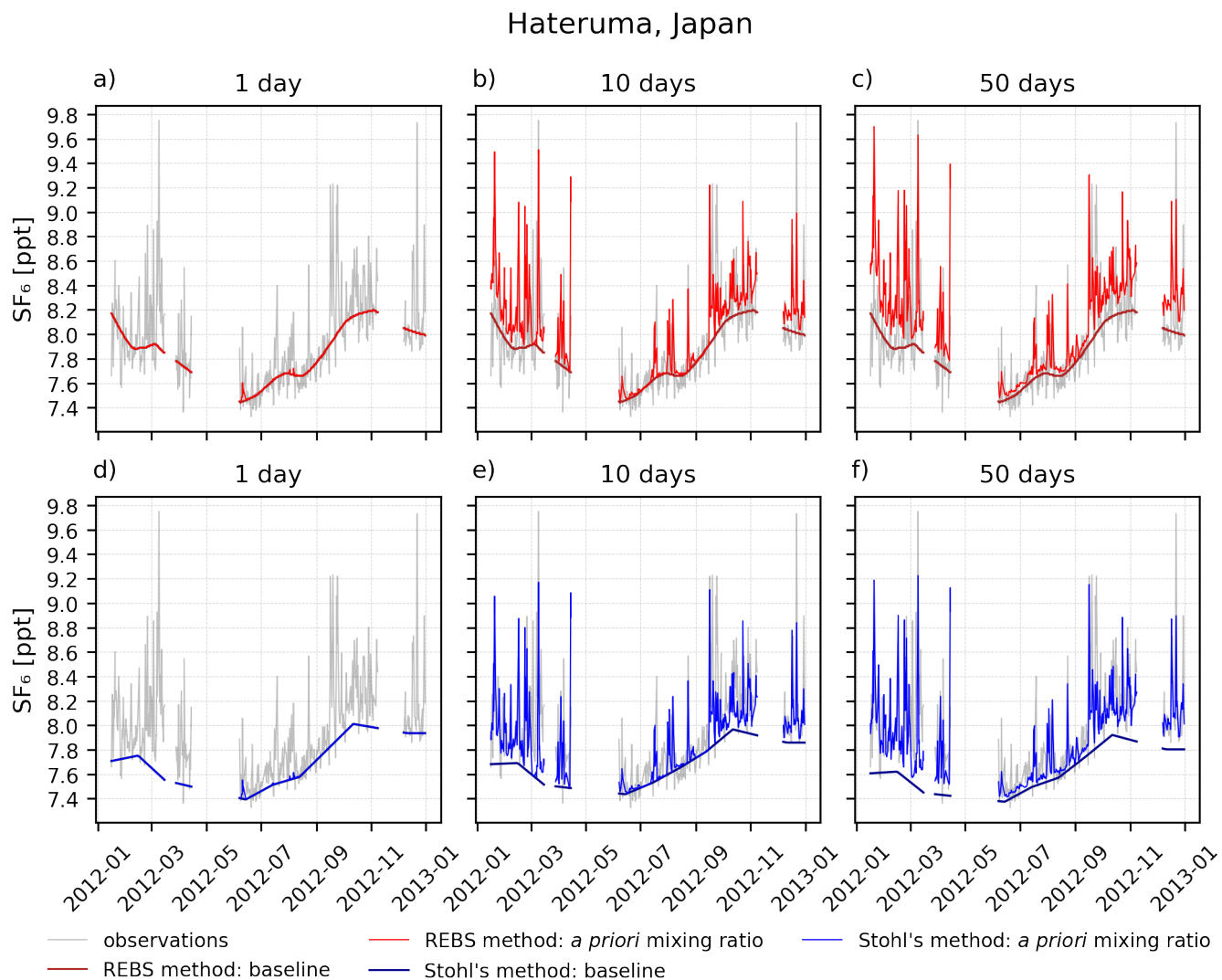


Figure S12: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Hateruma observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

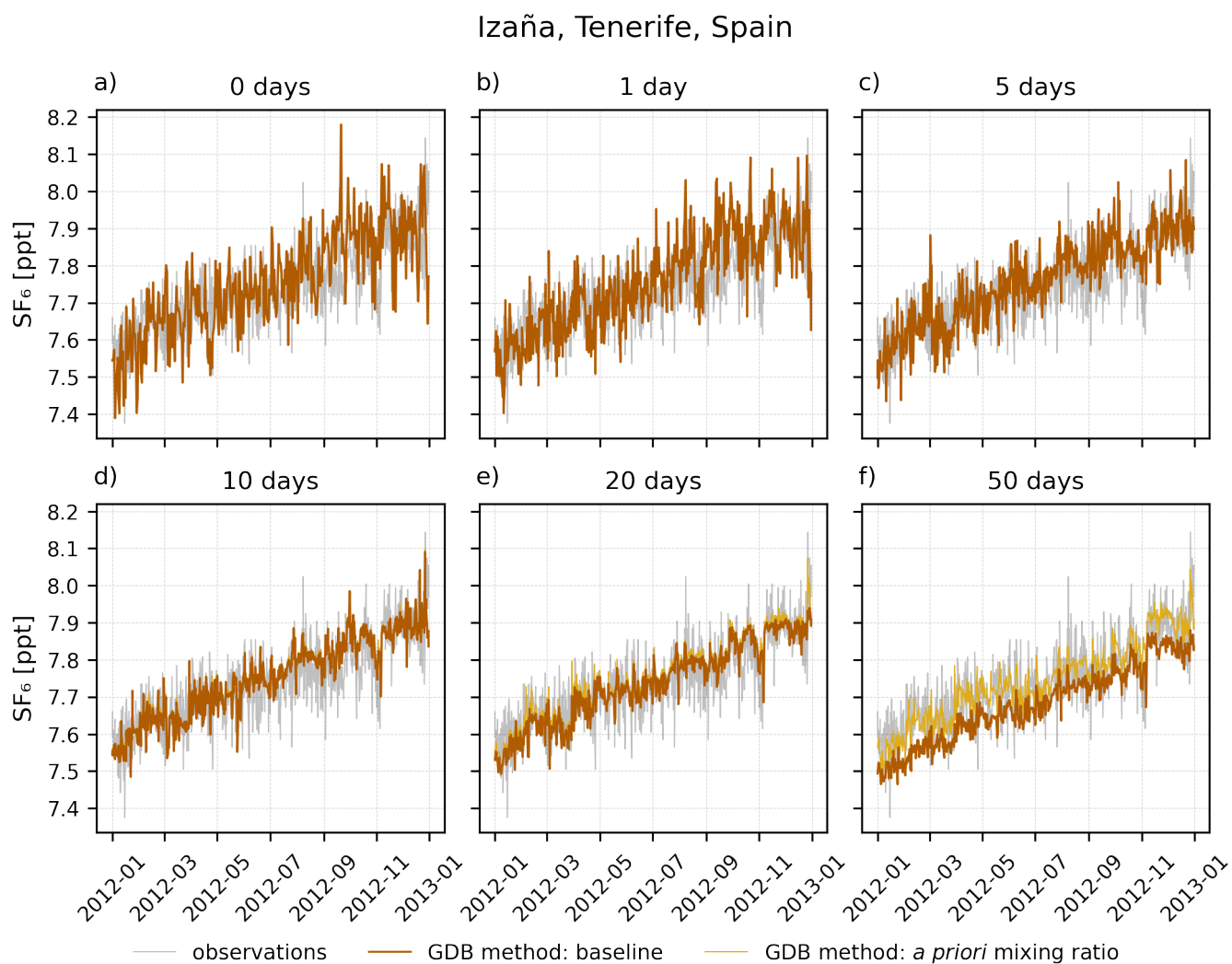


Figure S13: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Izaña observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

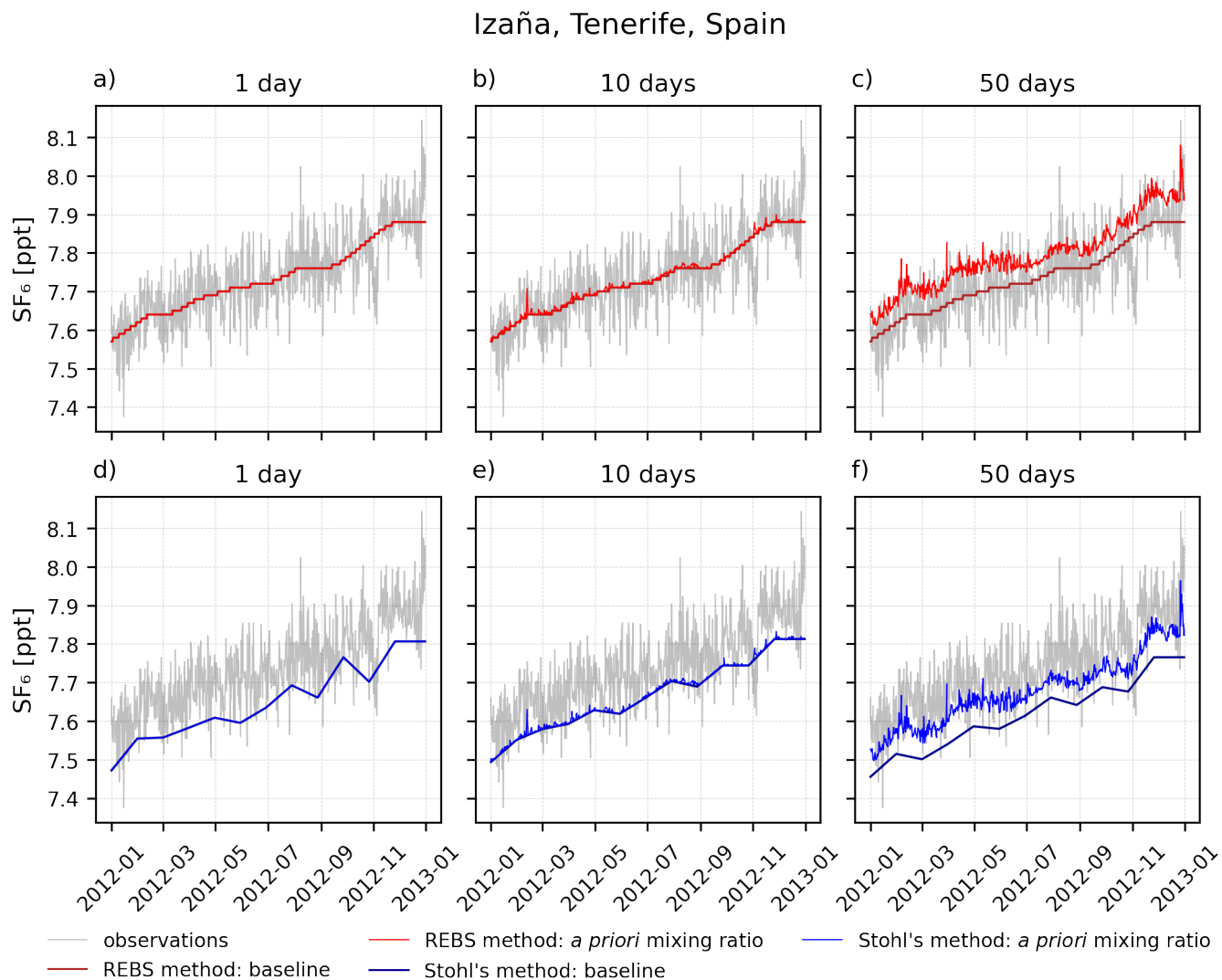


Figure S14: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Izaña observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

Jungfraujoch, Switzerland

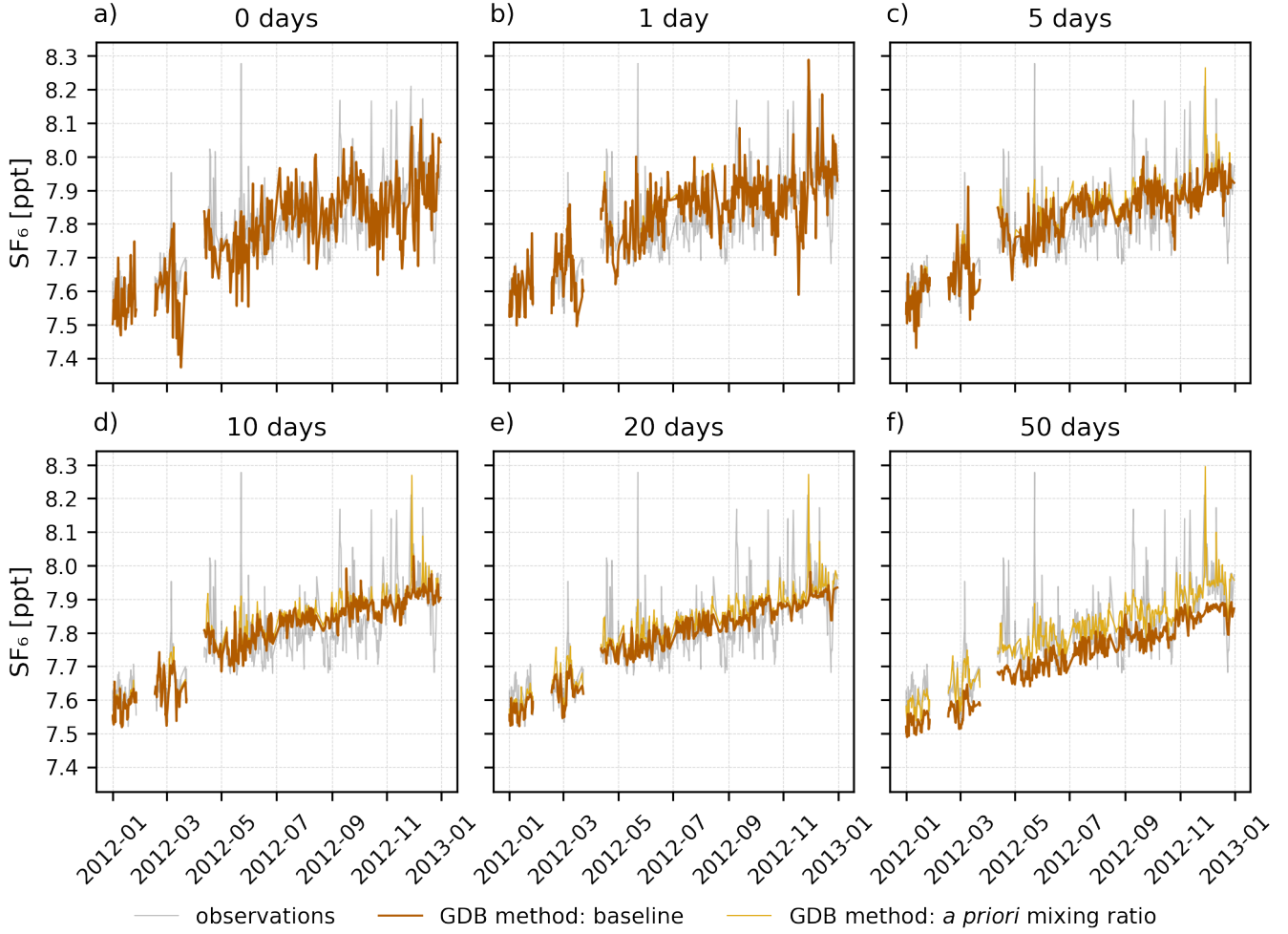


Figure S15: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Jungfraujoch observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

Jungfraujoch, Switzerland

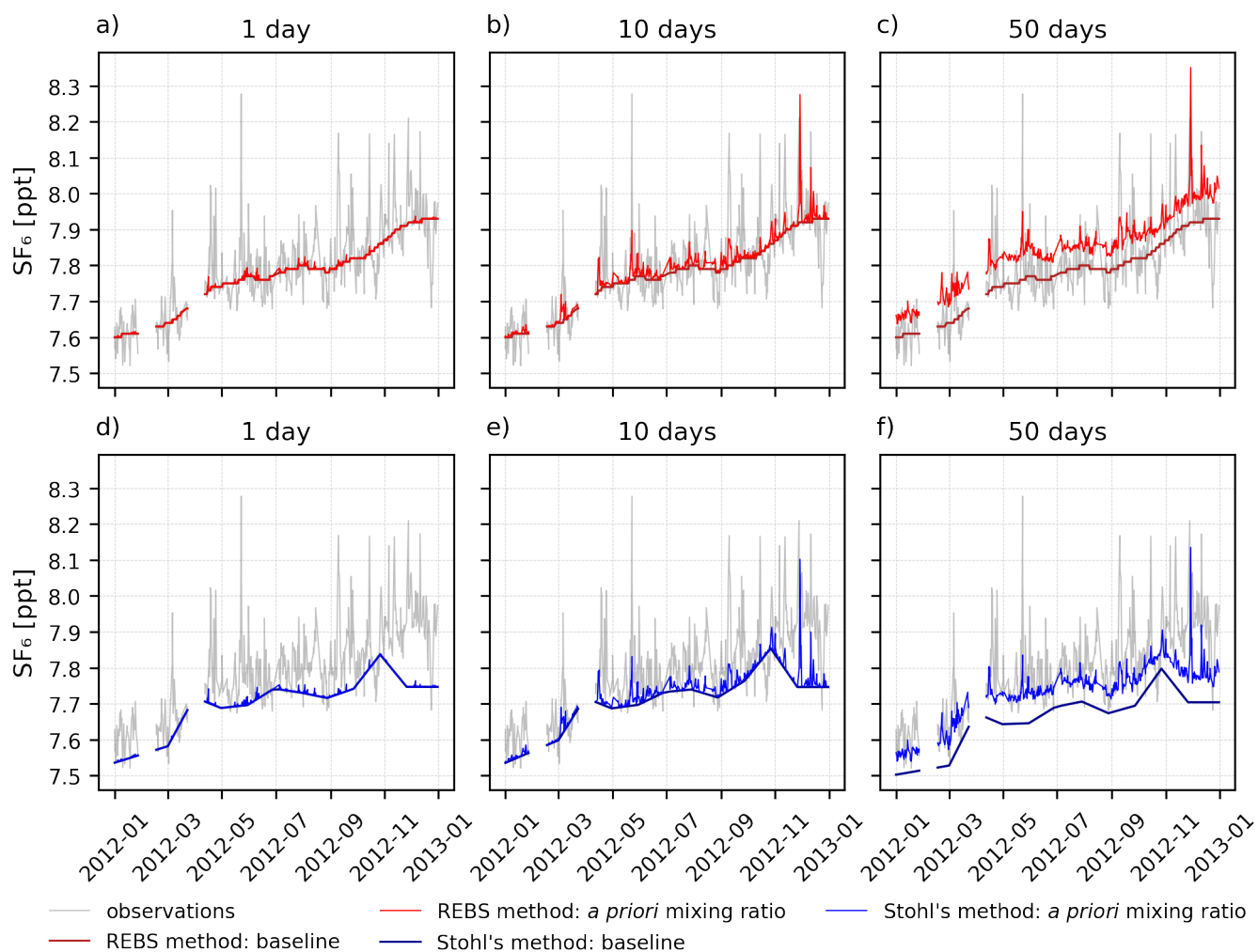


Figure S16: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Jungfraujoch observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

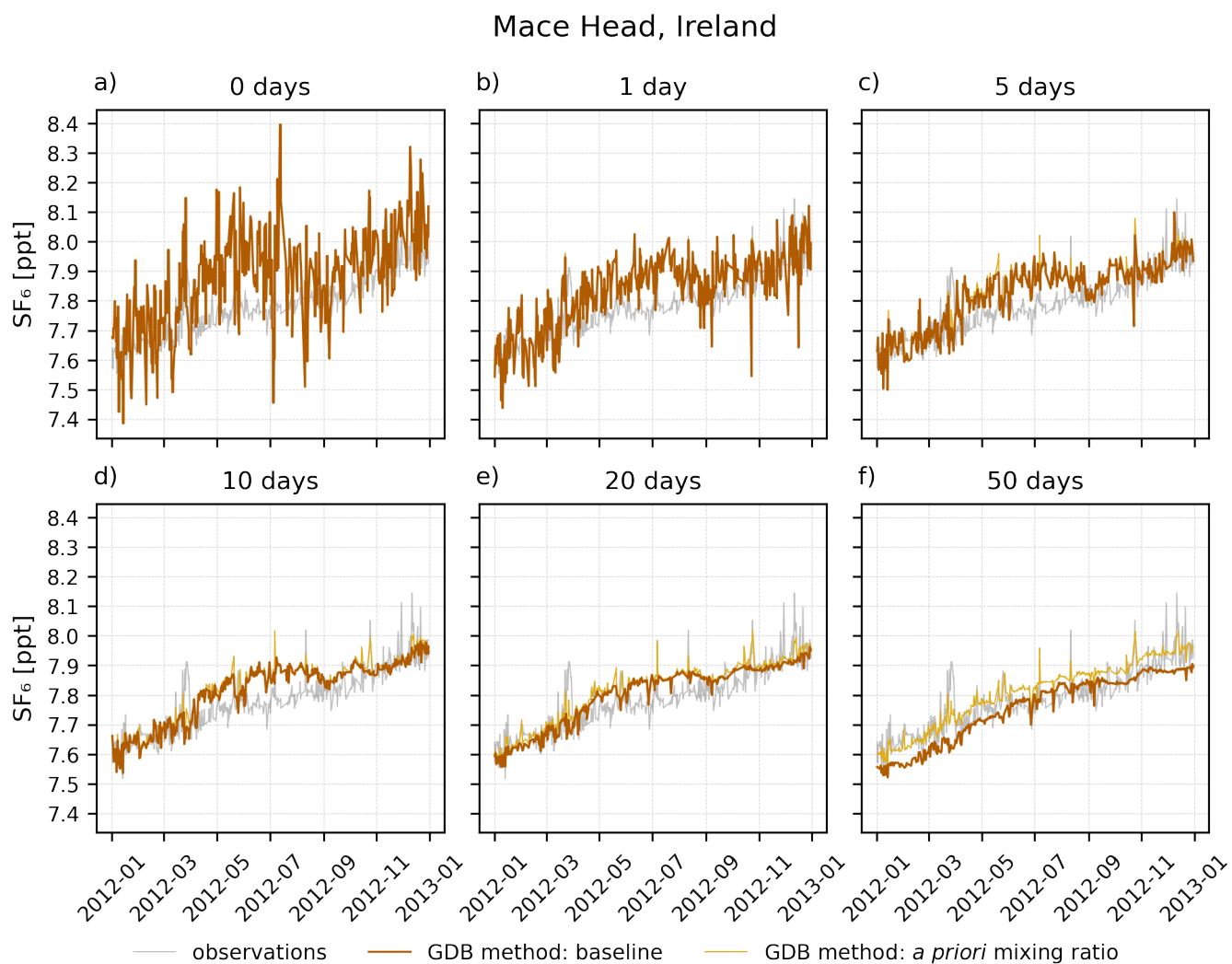


Figure S17: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Mace Head observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

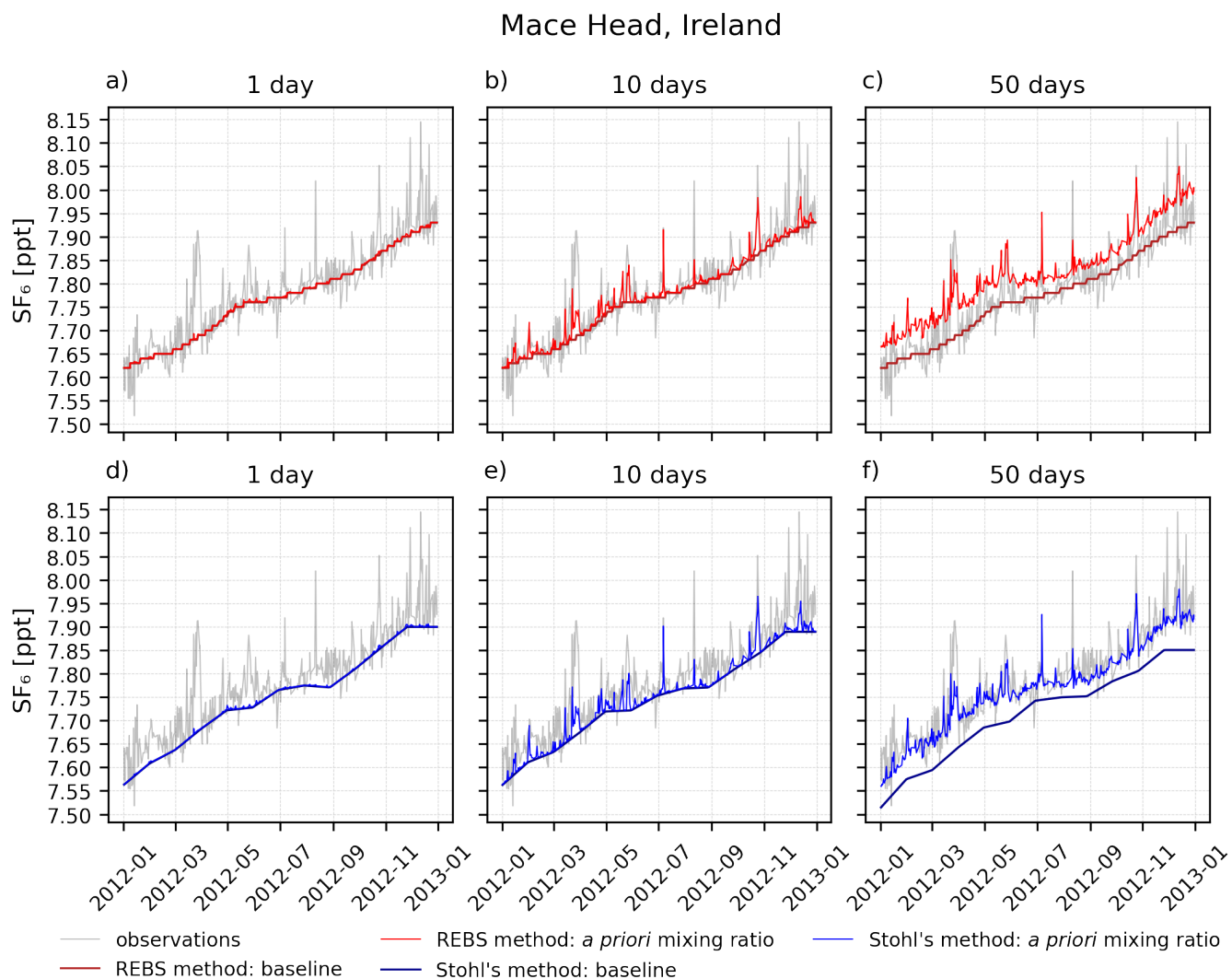


Figure S18: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Mace Head observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

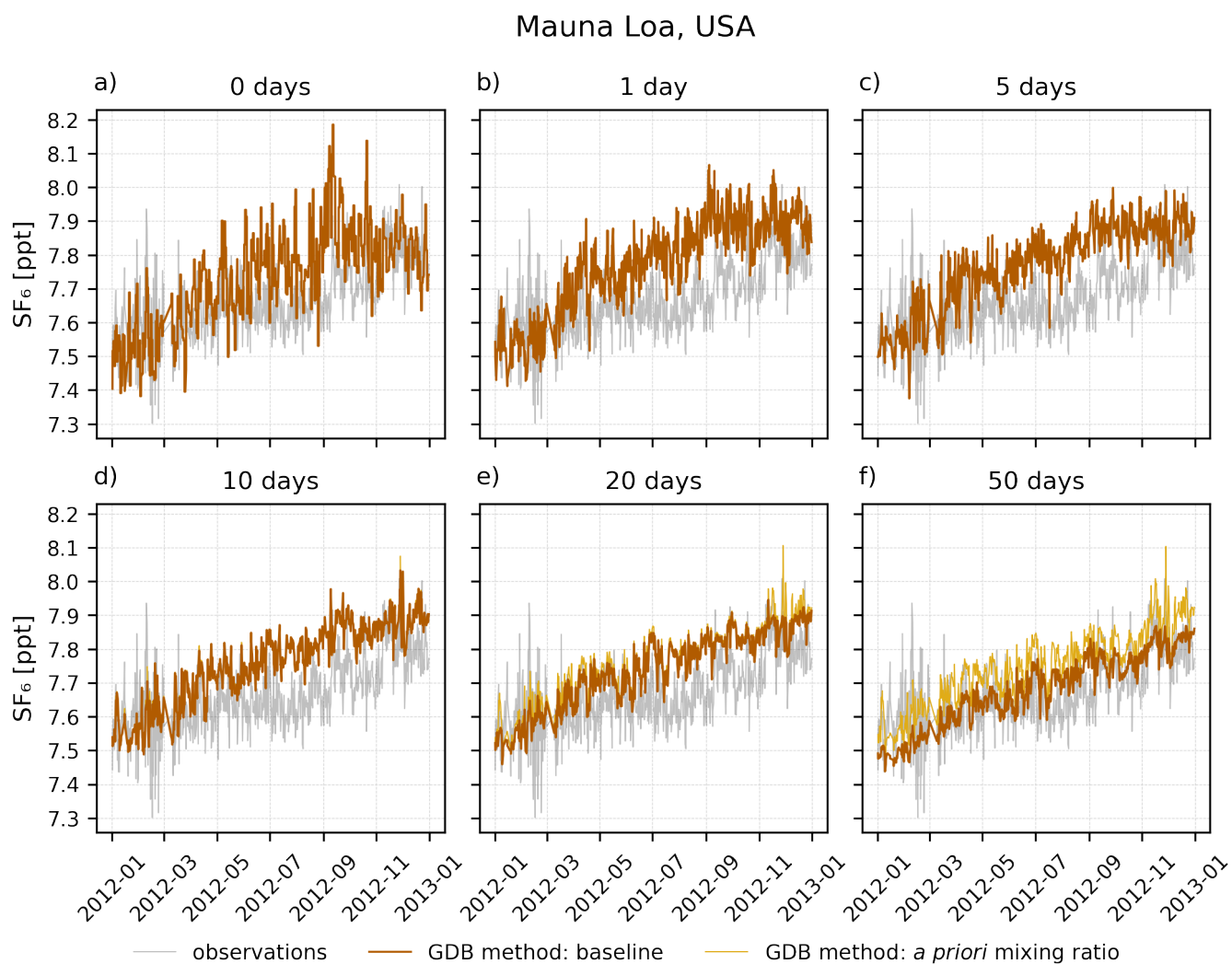


Figure S19: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Mauna Loa observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

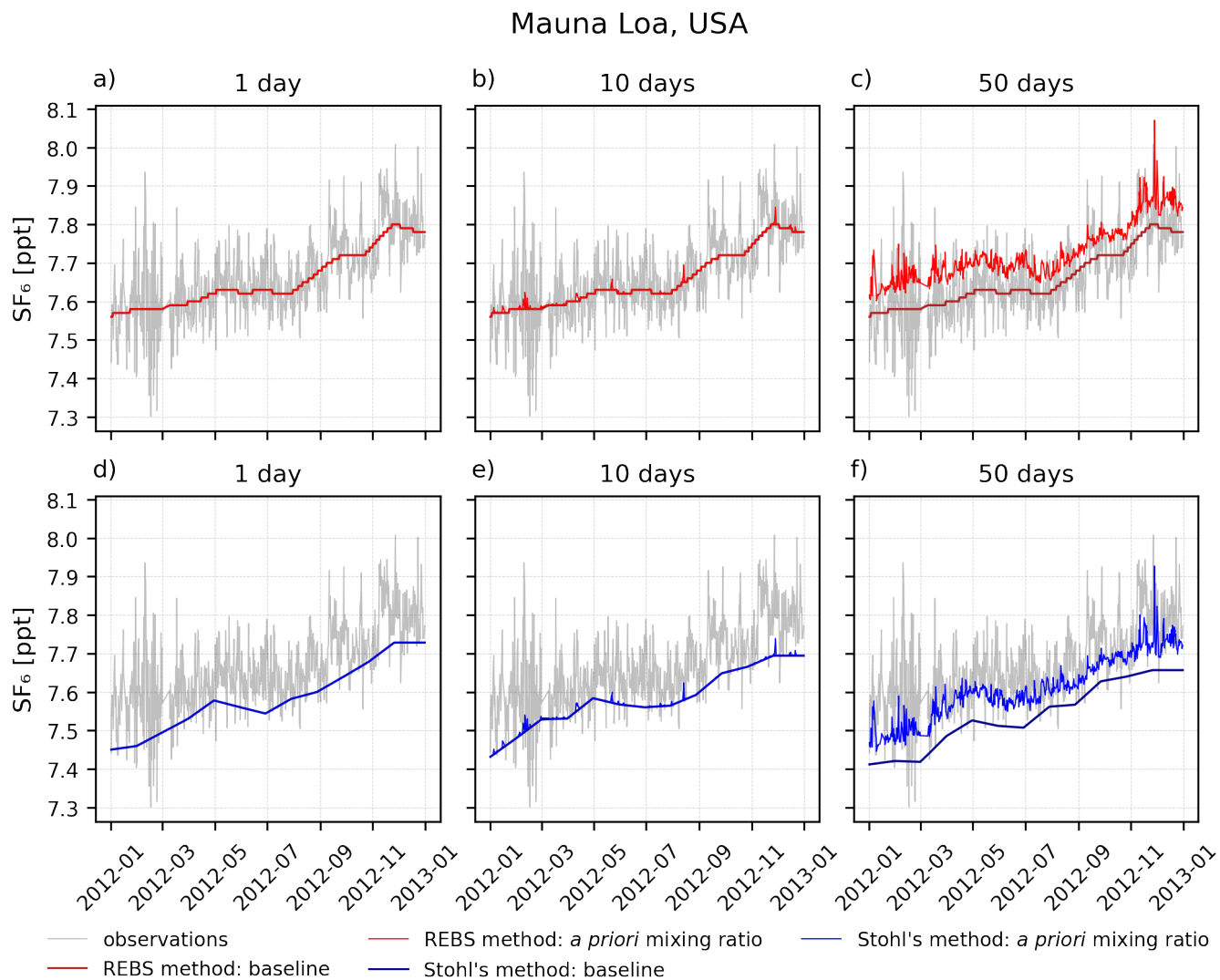


Figure S20: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Mauna Loa observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

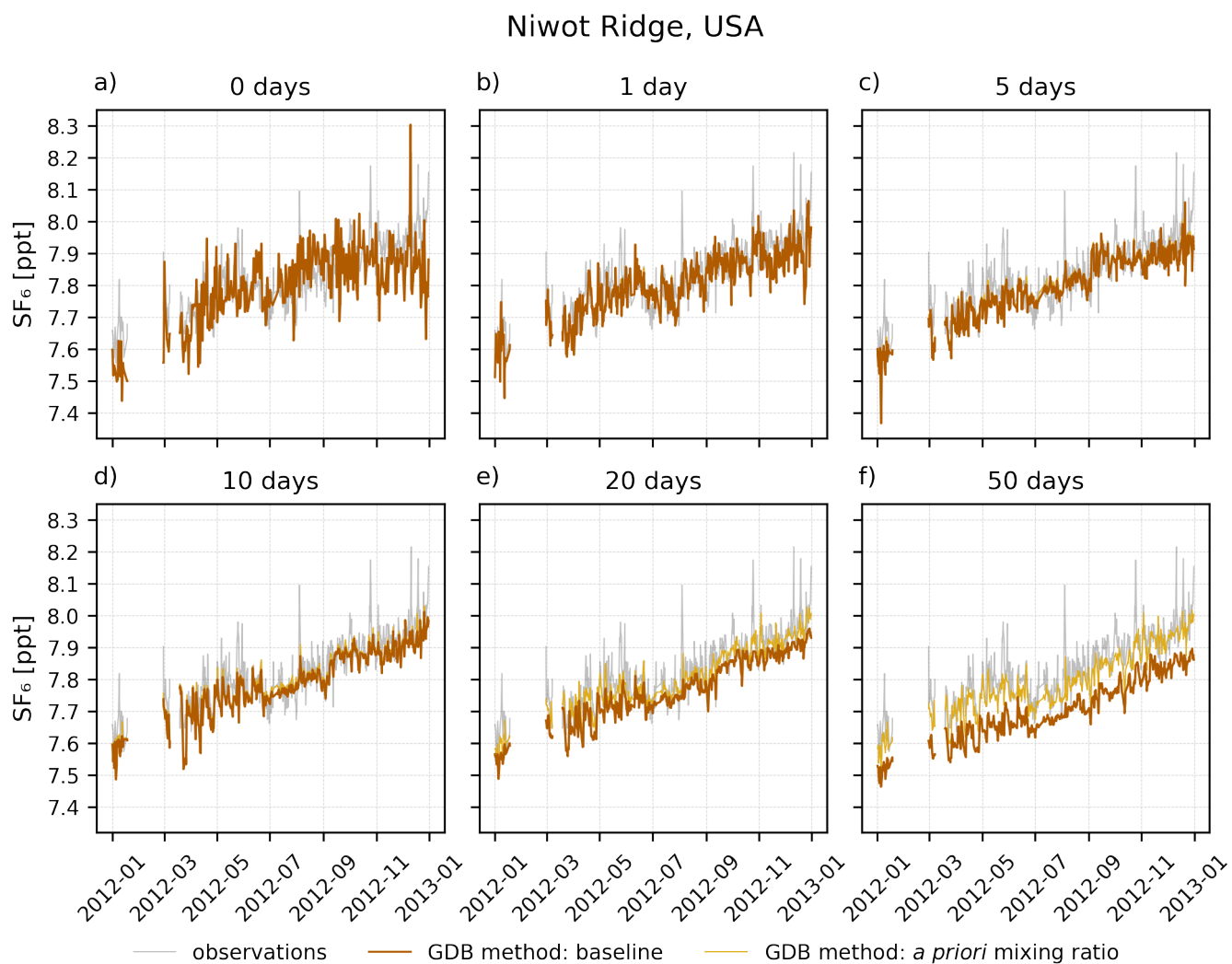


Figure S21: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Niwot Ridge observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

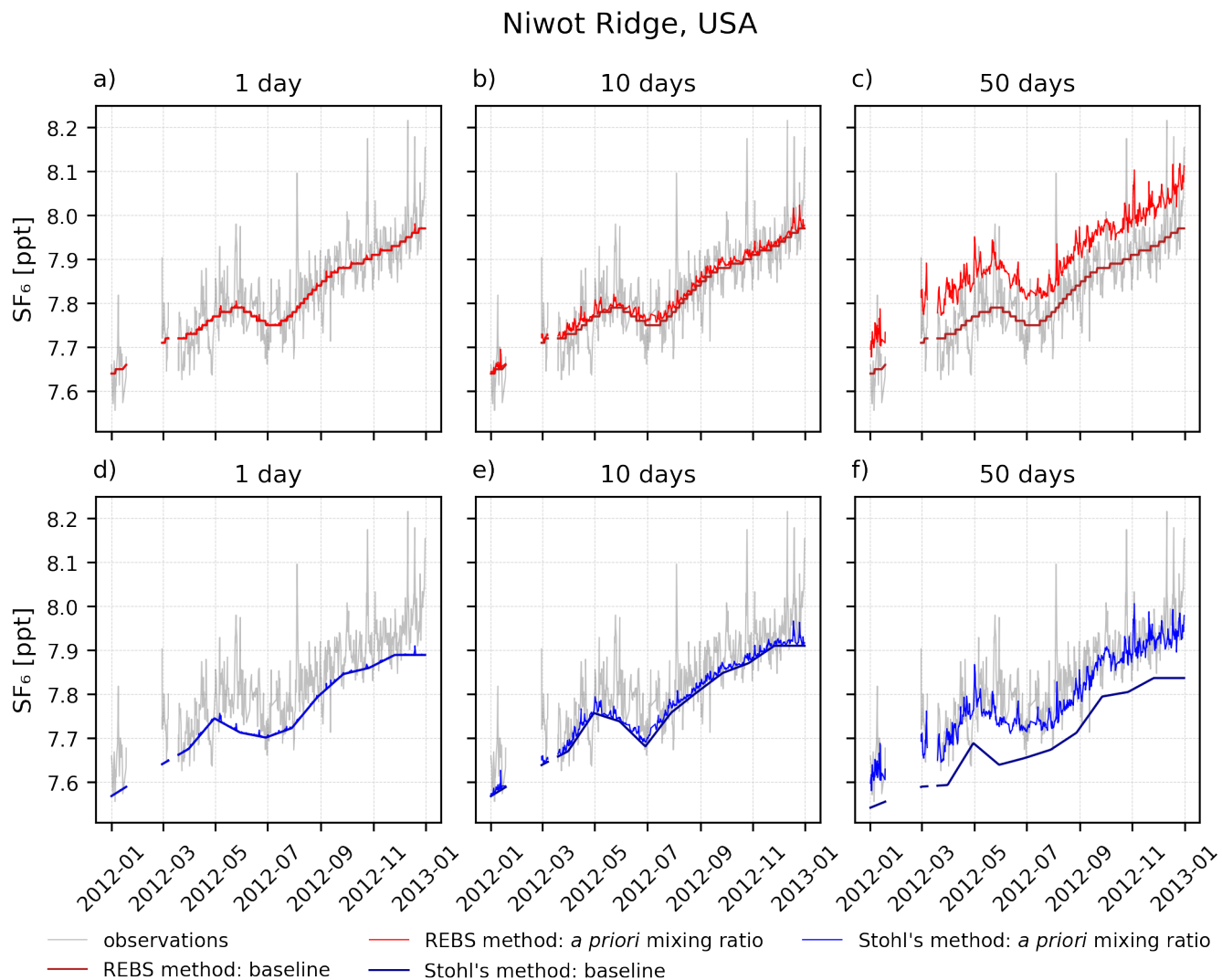


Figure S22: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Niwot Ridge observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

Ragged Point, Barbados

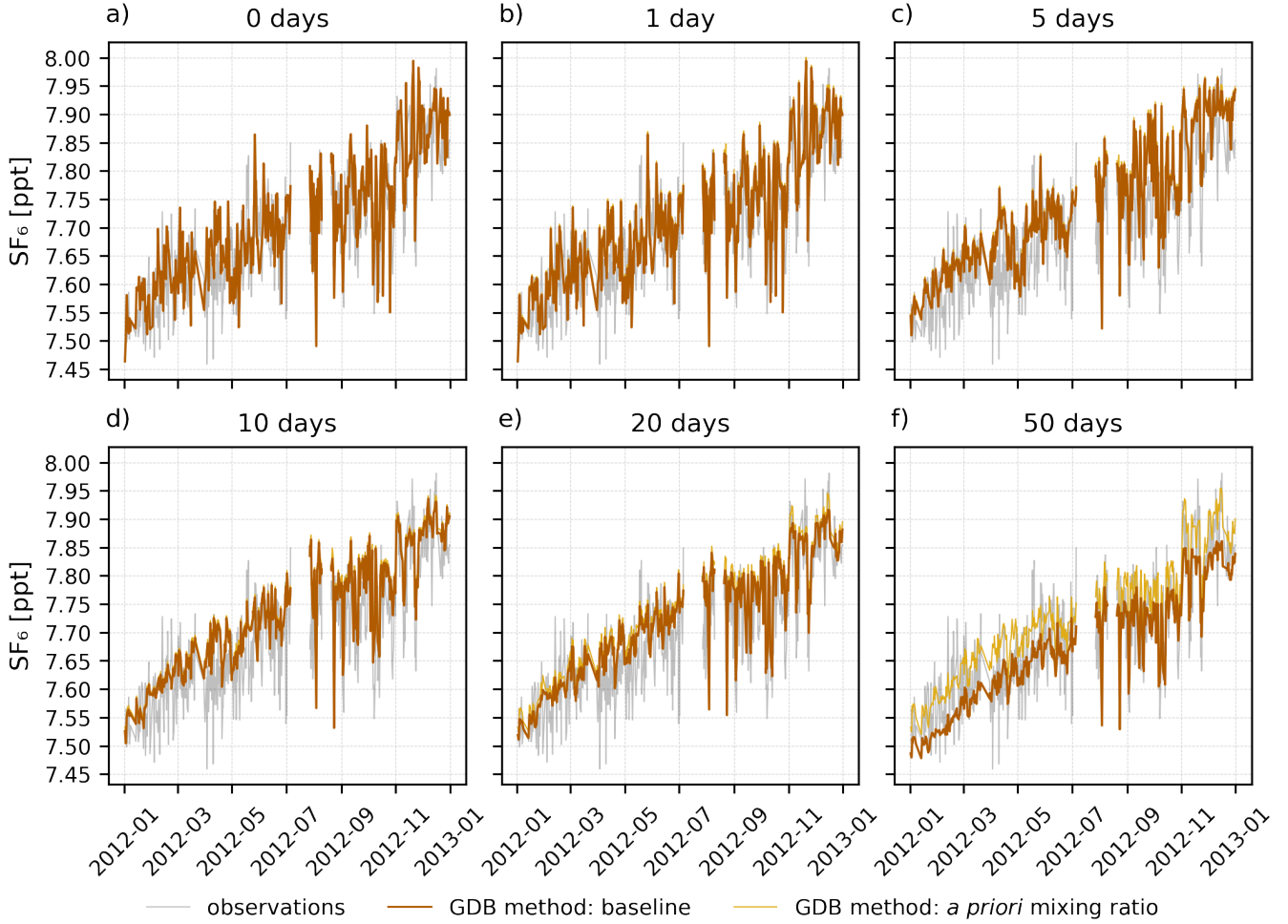


Figure S23: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Ragged Point observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

Ragged Point, Barbados

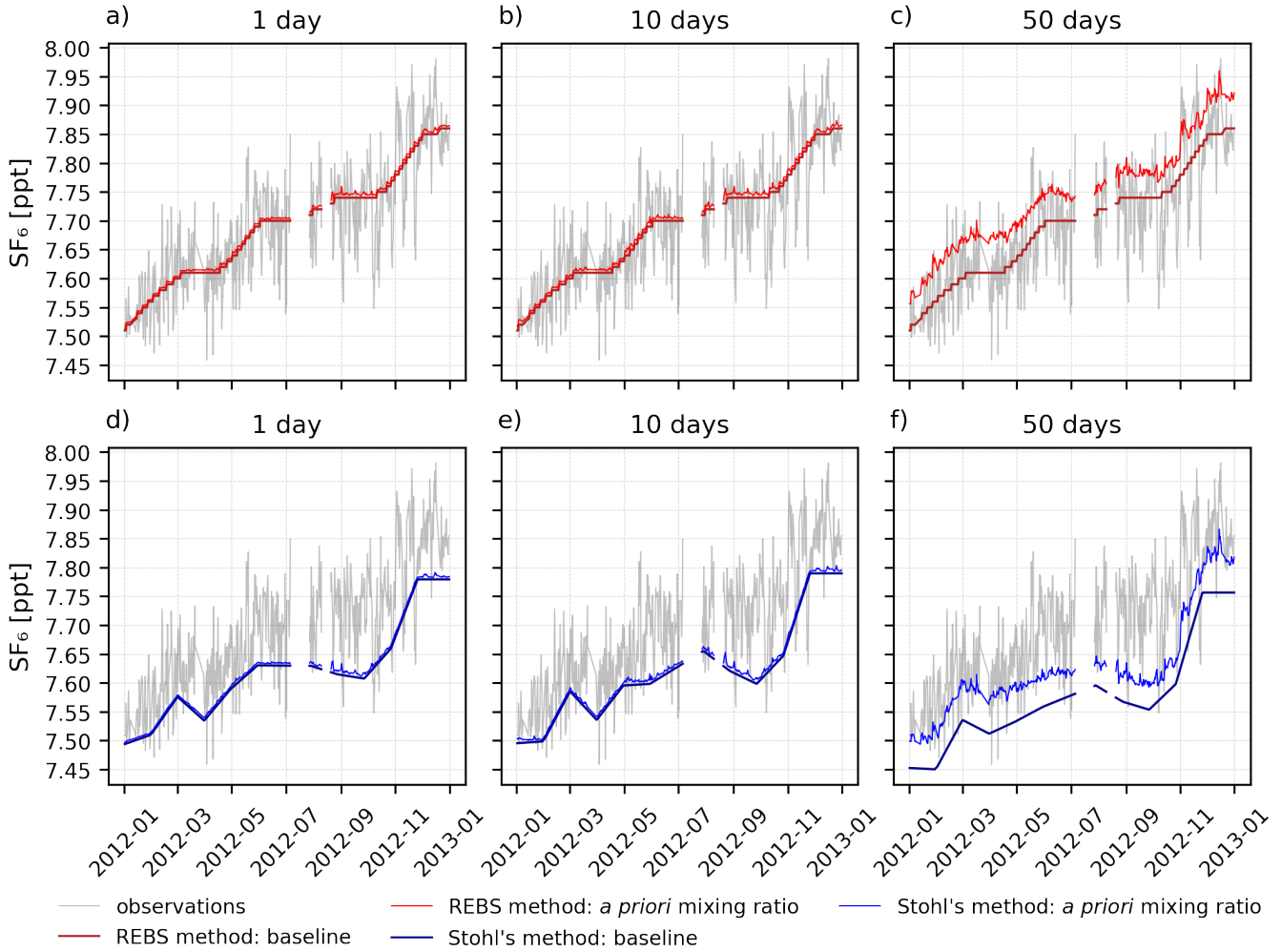


Figure S24: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Ragged Point observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

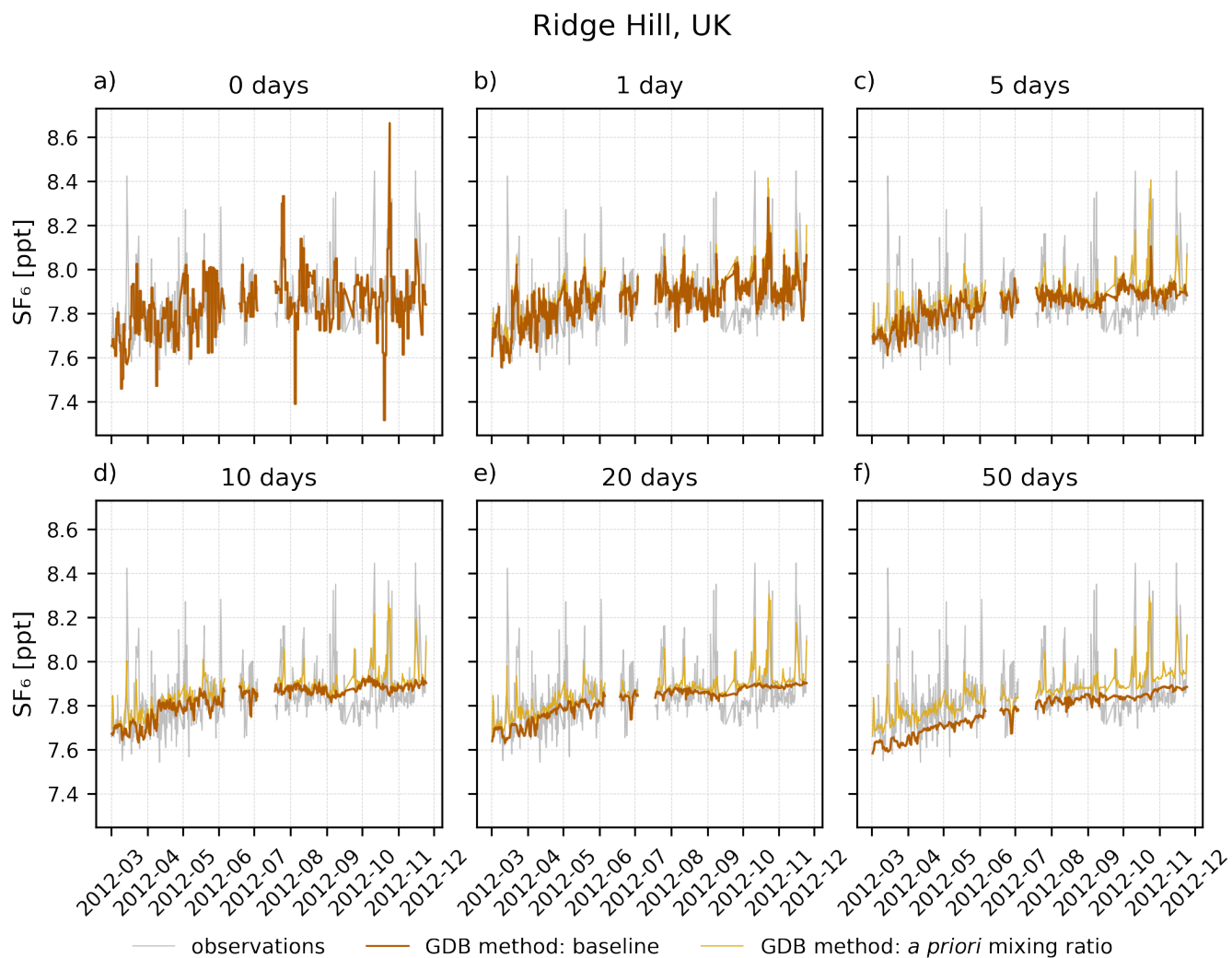


Figure S25: Baseline and *a priori* SF₆ mixing ratios calculated with the GDB method at the Ridge Hill observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

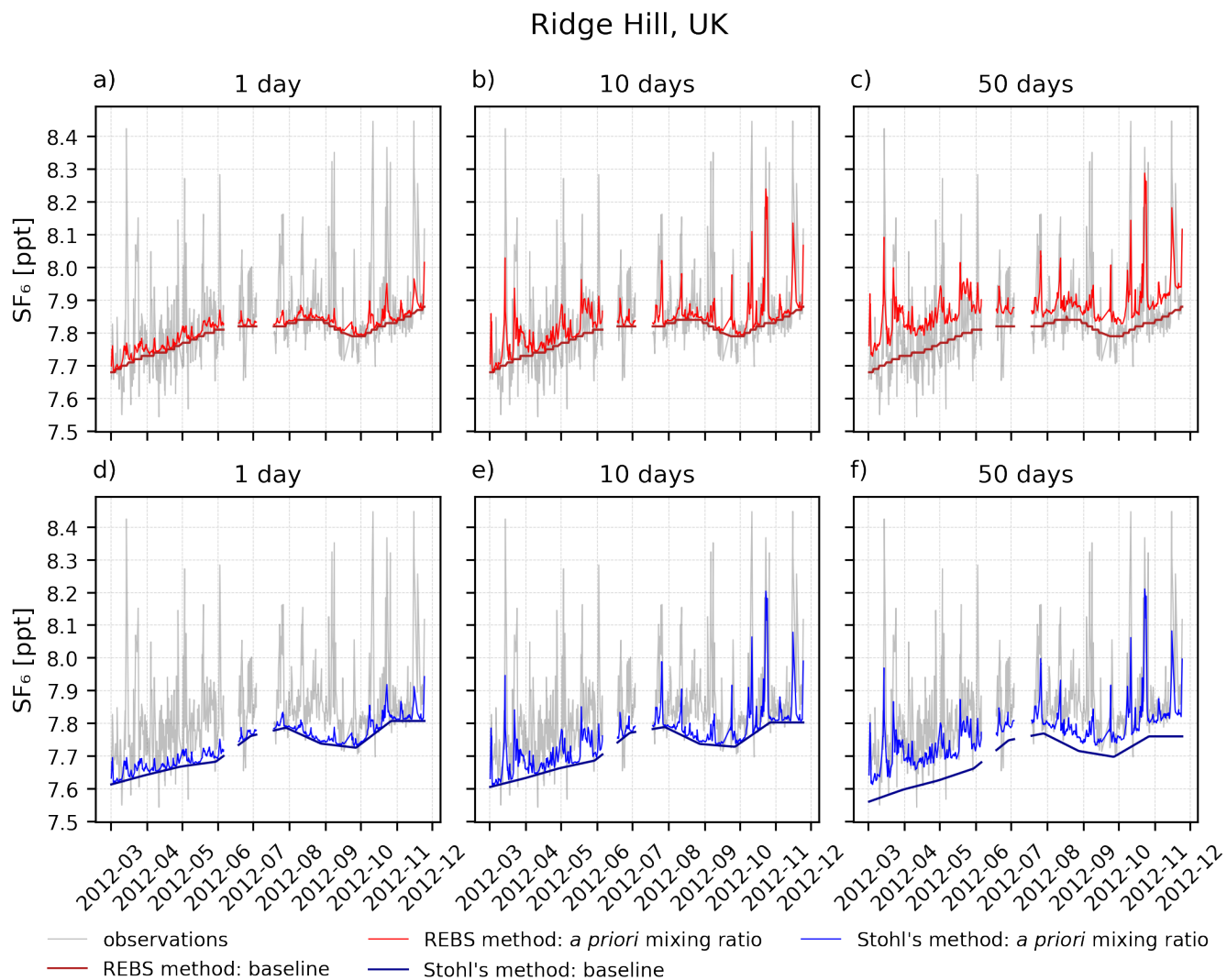


Figure S26: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Ridge Hill observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

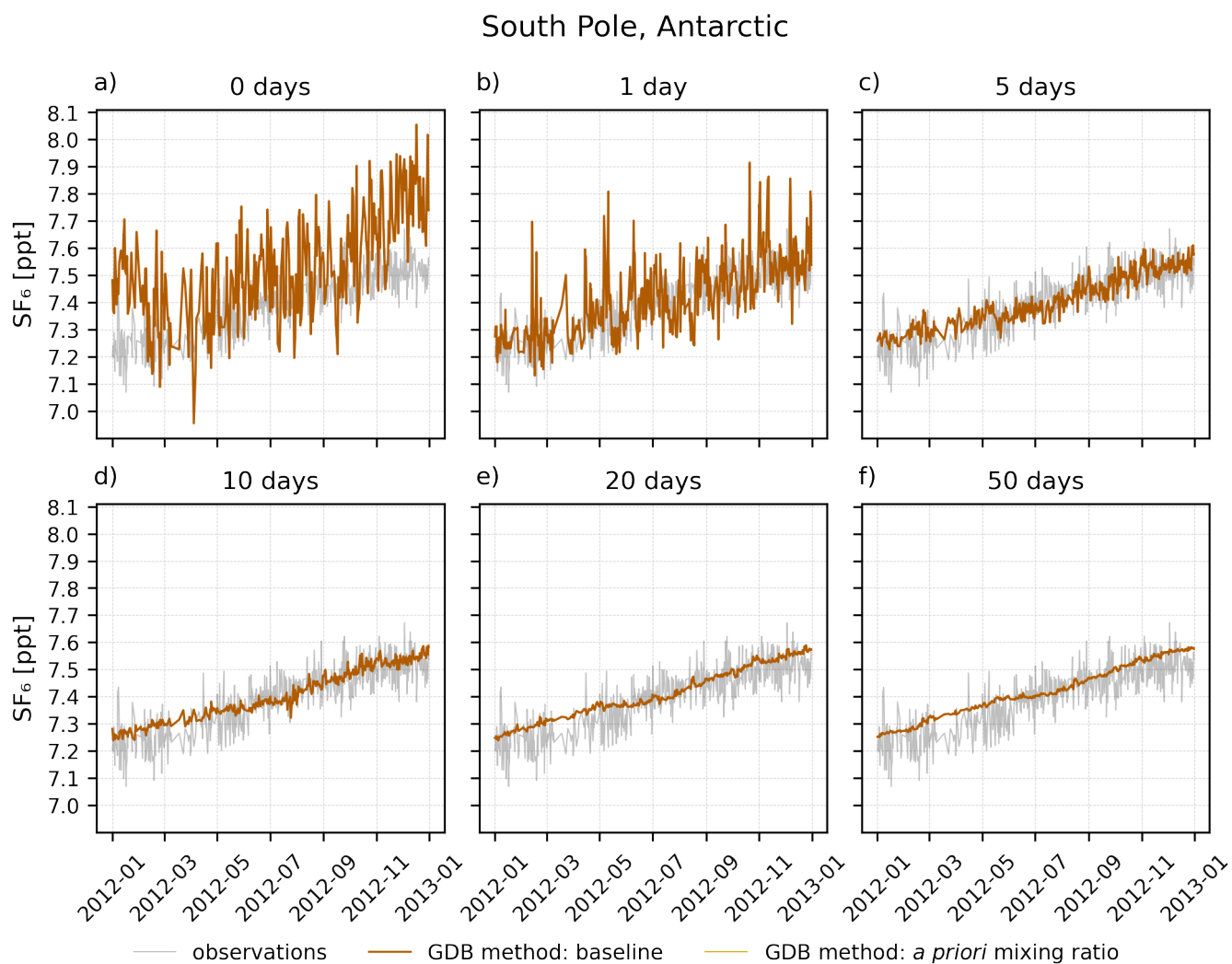


Figure S27: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the South Pole observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

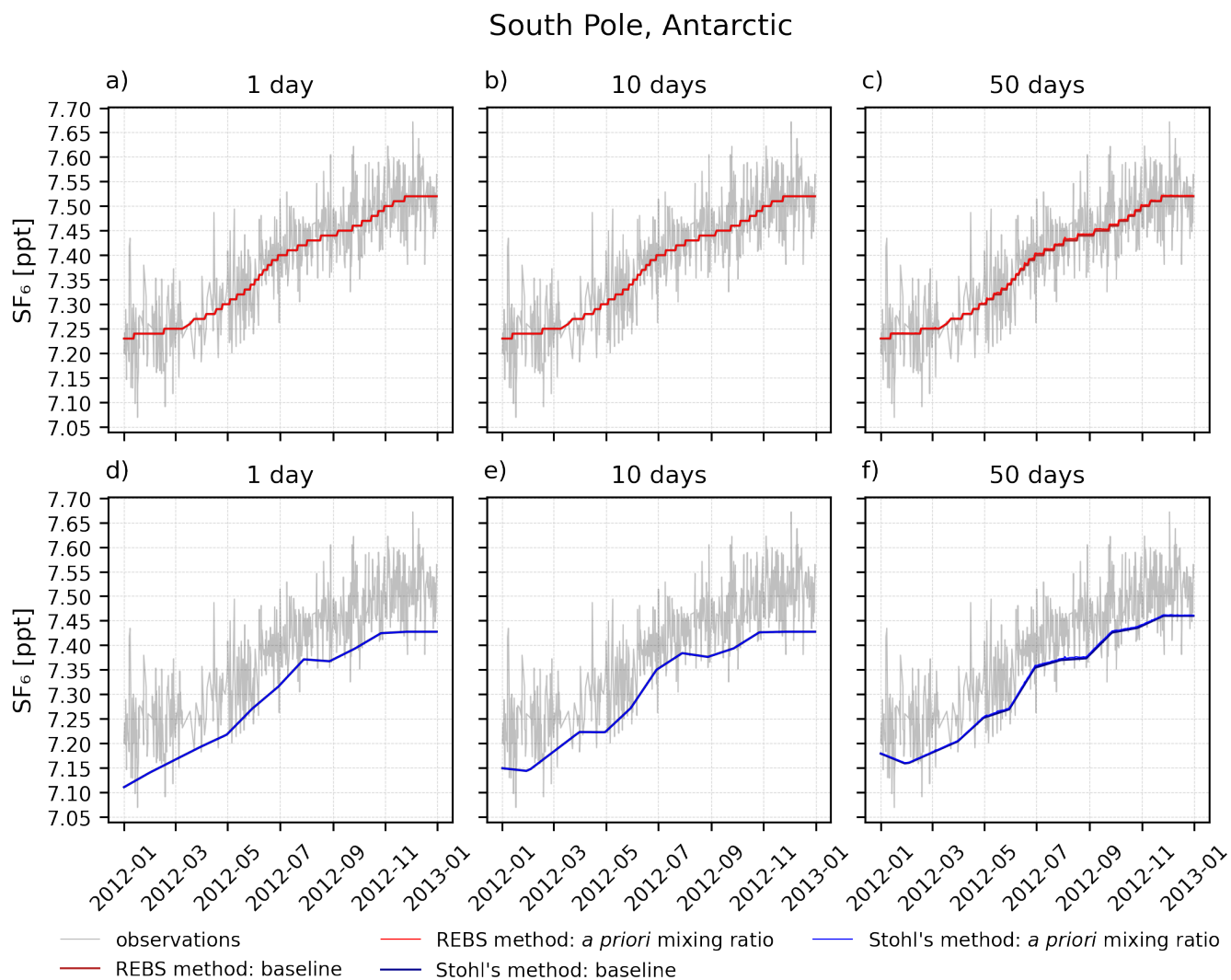


Figure S28: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the South Pole observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

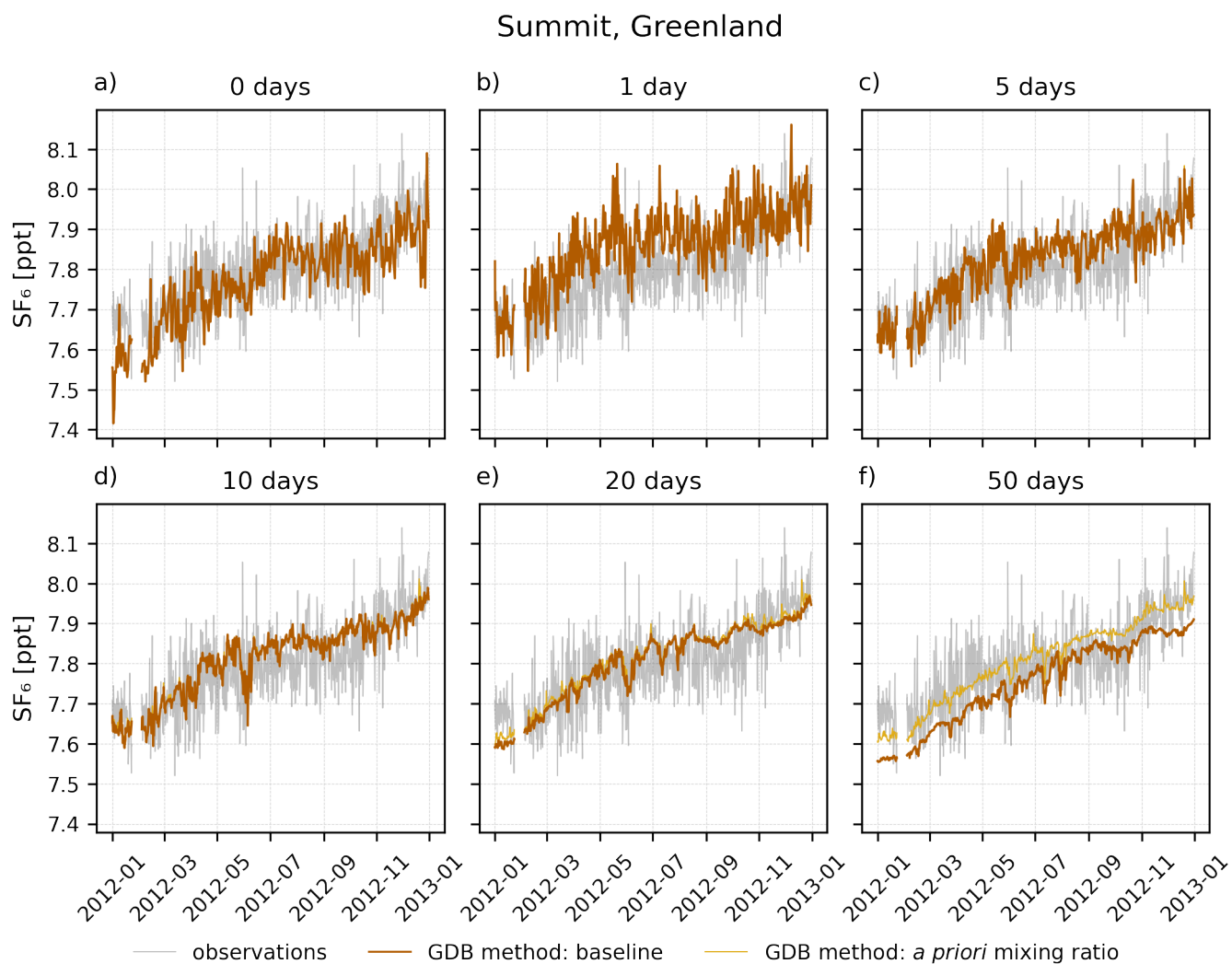


Figure S29: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Summit observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

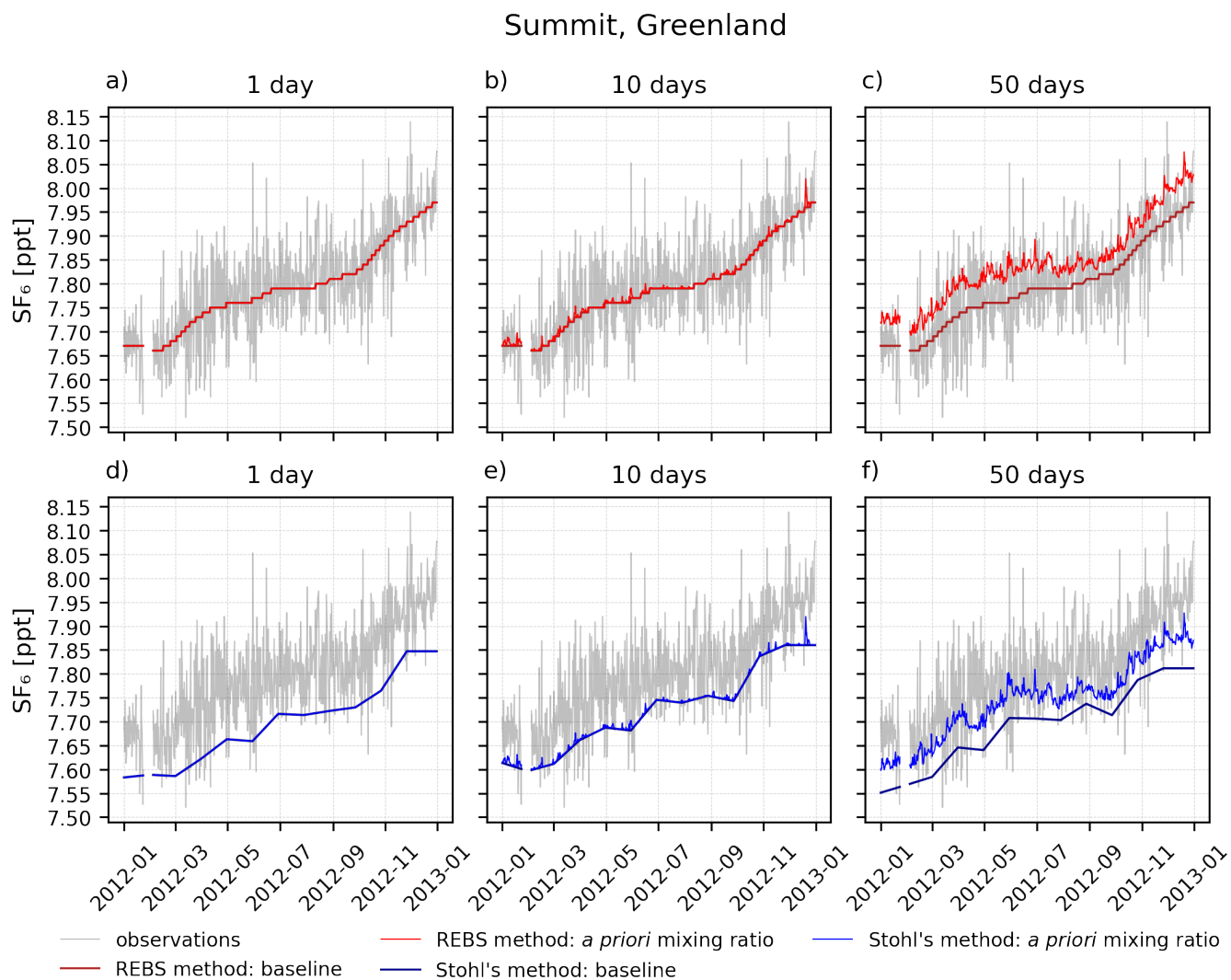


Figure S30: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Summit observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

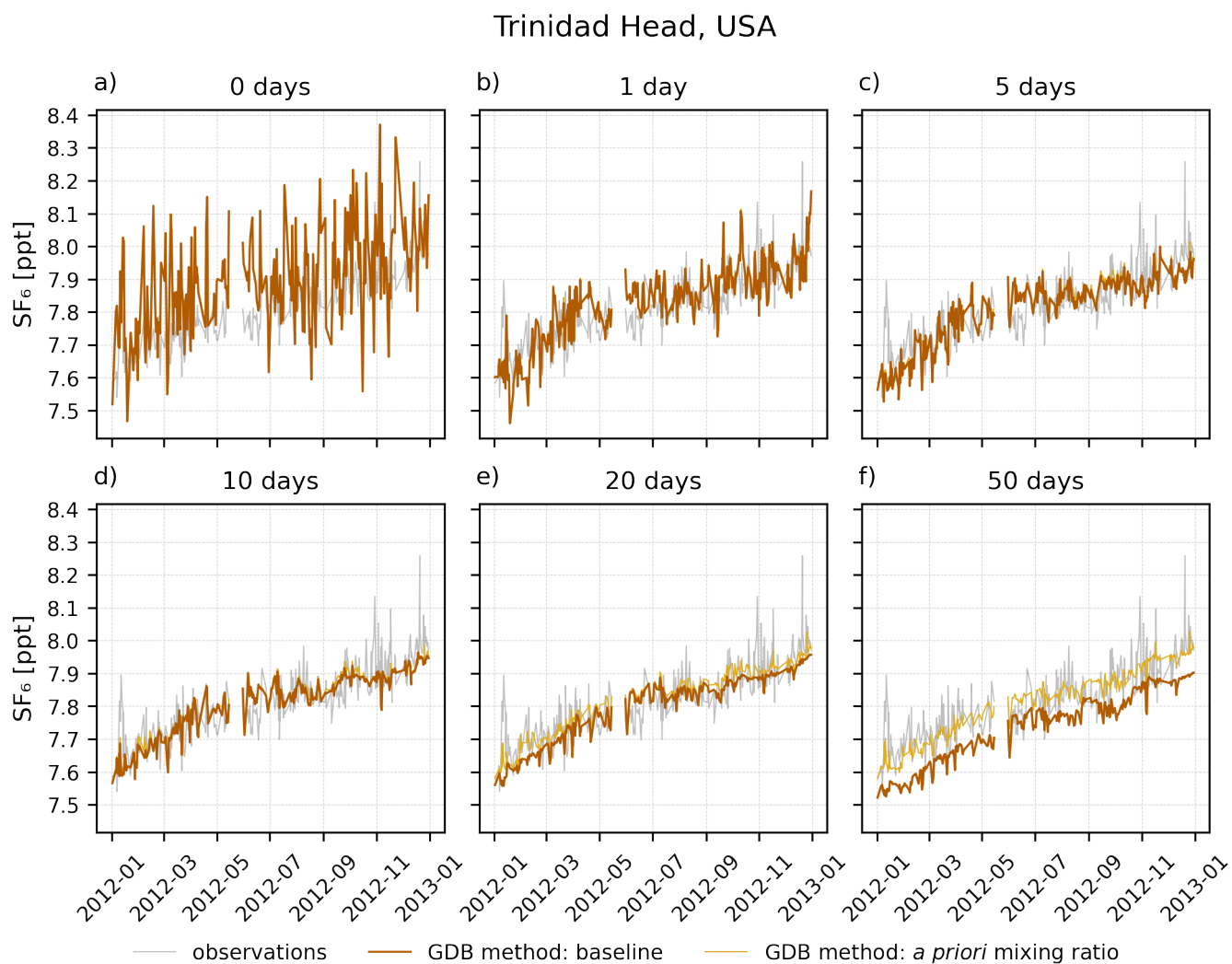


Figure S31: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Trinidad Head observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

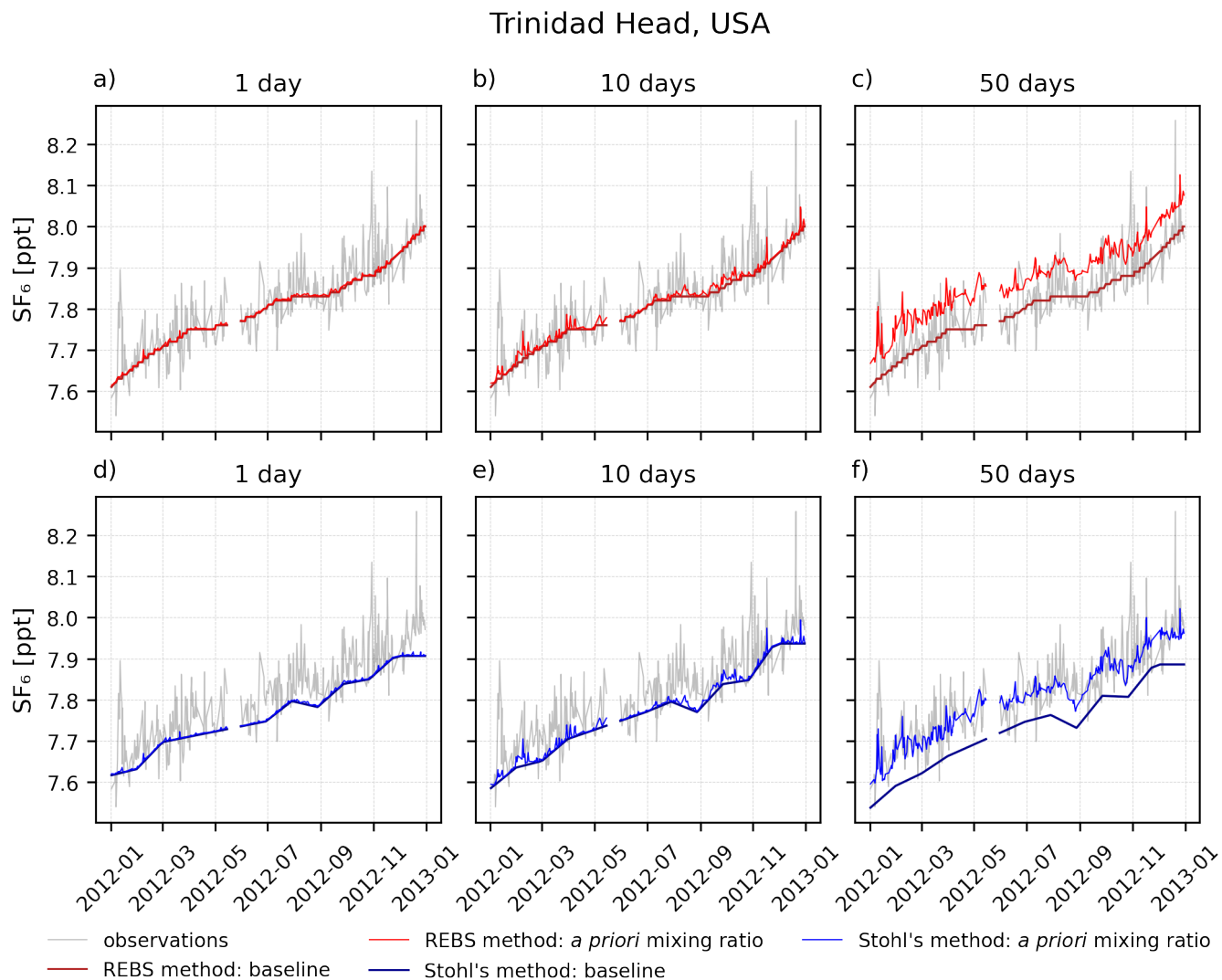


Figure S32: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Trinidad Head observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

Zeppelin, Ny-Alesund, Norway

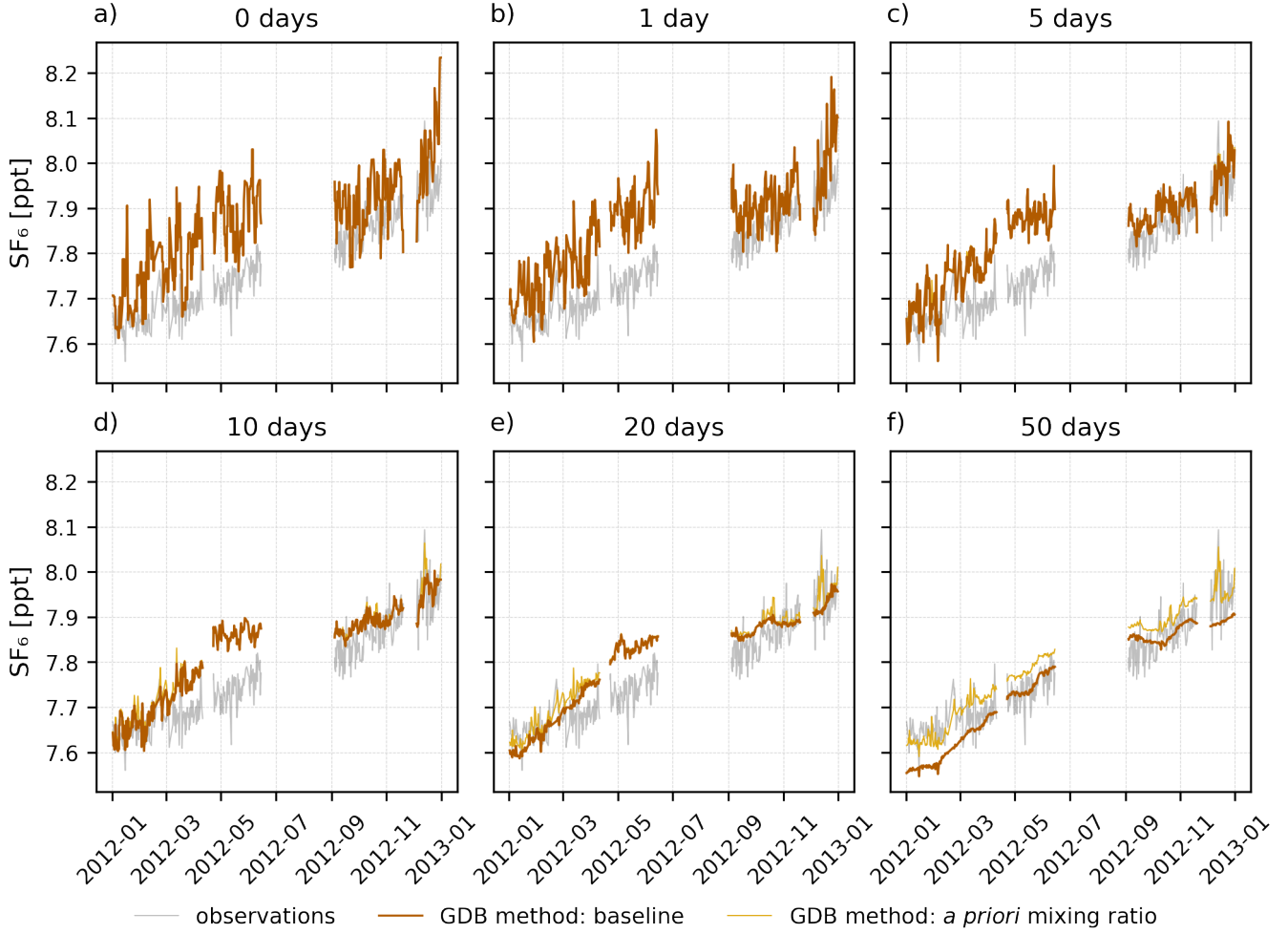


Figure S33: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Zeppelin observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

Zeppelin, Ny-Alesund, Norway

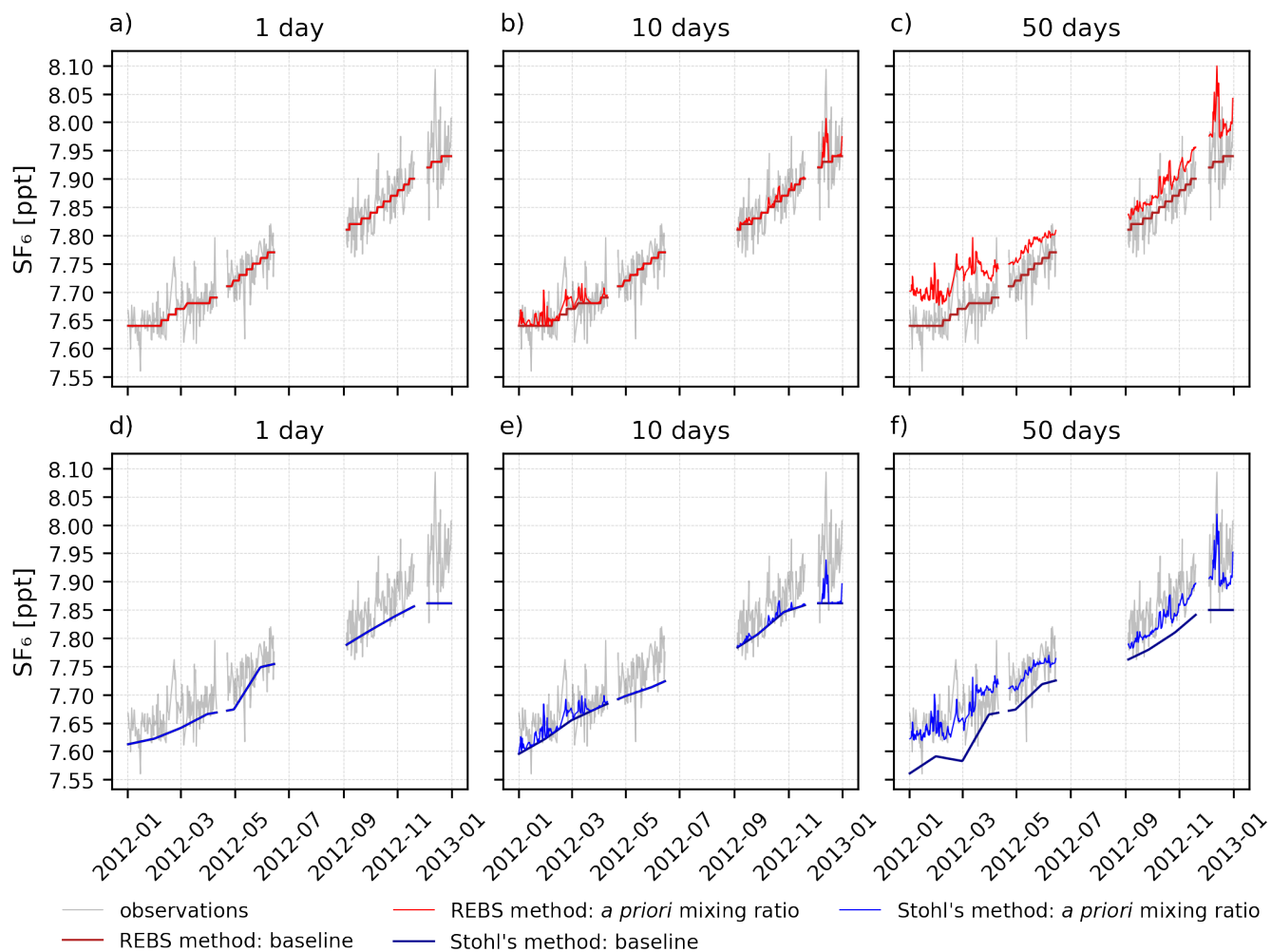


Figure S34: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Zeppelin observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).

Zugspitze-Schneefernerhaus, Germany

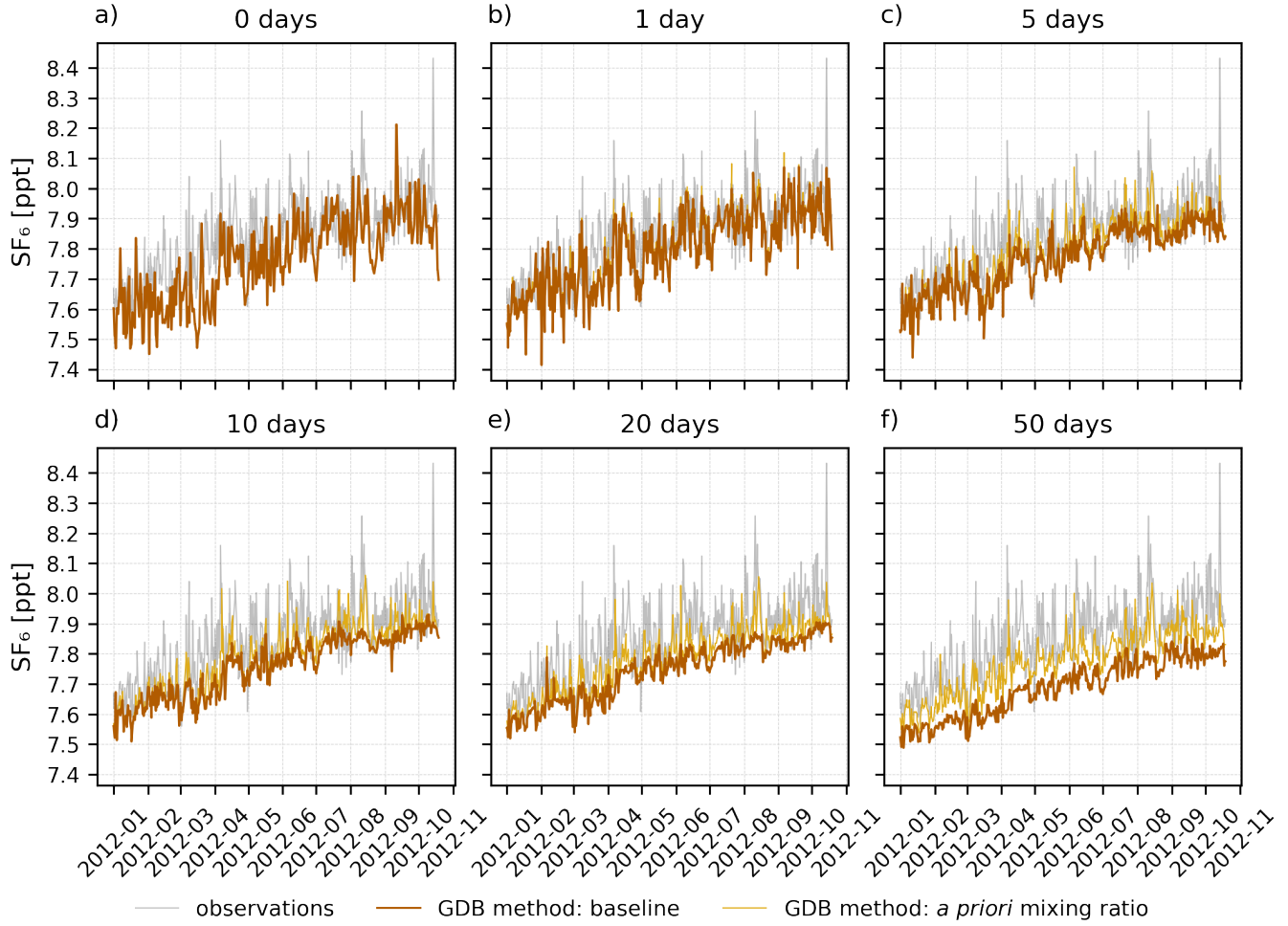


Figure S35: Baseline and *a priori* SF_6 mixing ratios calculated with the GDB method at the Zugspitze-Schneefernerhaus observation station for backward simulation times of 0 days (panel a), 1 day (b), 5 days (c), 10 days (d), 20 days (e) and 50 days (f).

Zugspitze-Schneefernerhaus, Germany

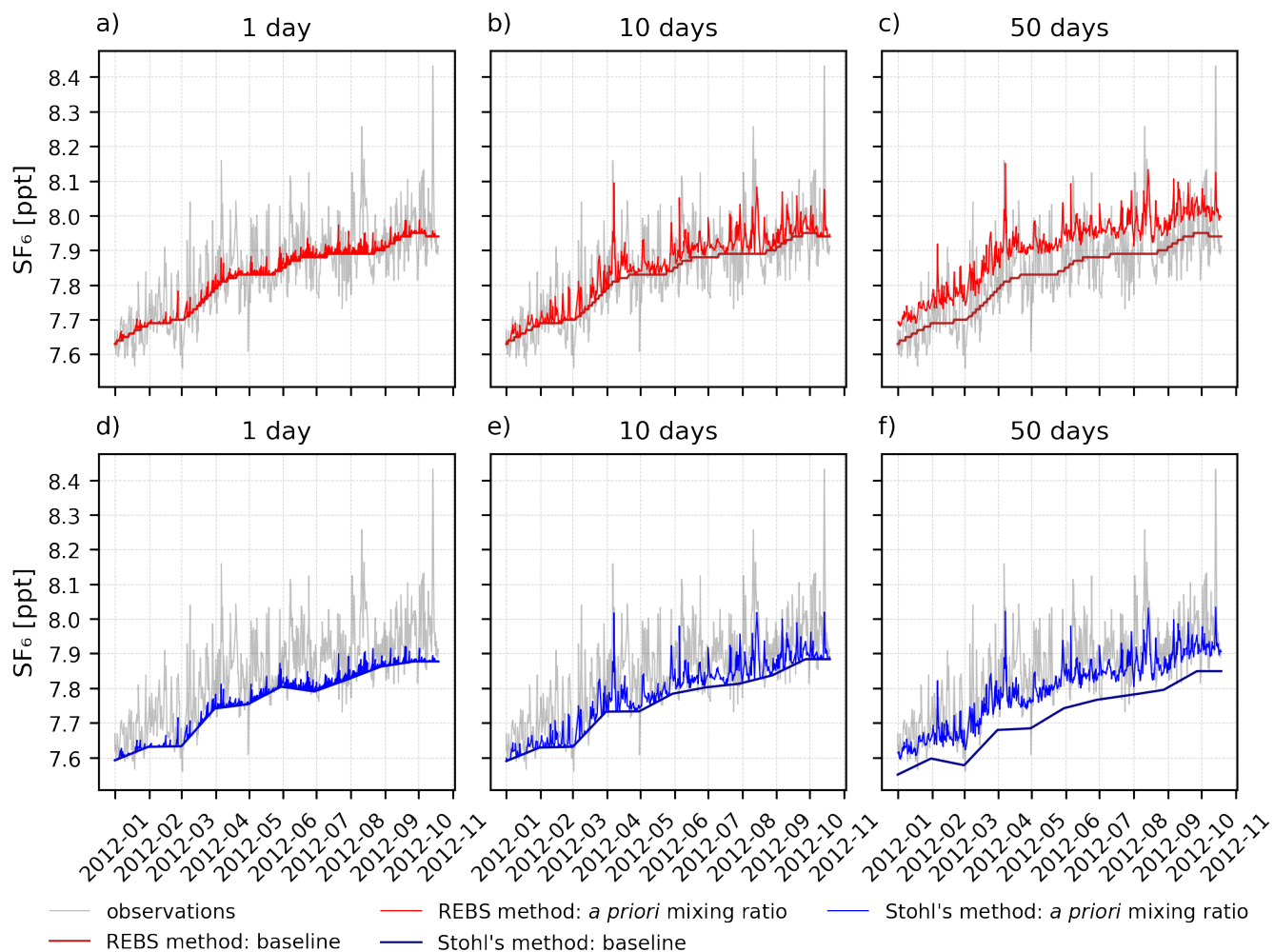


Figure S36: Baseline and *a priori* SF_6 mixing ratios calculated with the REBS (upper panels) and Stohl's method (lower panels) at the Zugspitze-Schneefernerhaus observation station, compared to SF_6 observations. Model results are shown for backward simulations of 1 day (panels a and d), 10 days (panels b and e) and 50 days (panels c and f).