

Appendix: Estimation of Retrieval Errors for SMILES Major Species

In this Appendix, the results of error estimations are shown in detail for all the 9 molecular species to be observed by SMILES, for ozone isotopes, and for ozone, H^{35}Cl , and HO_2 in the mesosphere, using standard atmospheric profiles in the polar, mid-latitude, and equatorial regions.

A.1 Radical Observation

Simulations are set up as follows. The spectroscopic line parameters, such as line position and line strength, are taken from the JPL catalog, and pressure broadening parameters are given by the HITRAN catalog. The height profiles of molecular volume mixing ratios, temperature, and pressure used in the simulations are the standard profiles for the mid-latitude, equatorial, and polar regions given by the Rutherford Appleton Laboratory.

For the instrumental parameters, representative values of the SMILES instrument are used. The retrieval error in the volume mixing ratio, the error ratio defined as the ratio of the retrieval error to the *a priori* error, and the averaging kernels are calculated for each molecules in the three SMILES observation bands. The frequency grid for the forward model is 0.8 MHz, and no frequency binning is applied for the retrieval. The molecules considered in the forward model and retrieval are normal O_3 , ozone isotopes, vibrationally-excited O_3 , HCl , ClO , HOCl , H_2O_2 , HO_2 , HNO_3 , BrO , and CH_3CN , as well as H_2O , N_2 , and O_2 . The *a priori* profiles are assumed to coincide with the true profiles for all the molecular species except ozone isotopes. The *a priori* standard deviations are assumed to be 100 percent of the *a priori* profiles for the research products and 50 percent for the standard products. The measurement error covariance matrix \mathbf{S}_ϵ is given from the measurement noise, $\sigma = (T_{sys} + T_{atm})/\sqrt{B\tau}$, where T_{sys} is the receiver system noise temperature assumed to be 500 K, and T_{atm} is the antenna temperature from the spectrum, B is the equivalent noise bandwidth assumed to be 0.8 MHz, τ is the integration time. The integration time τ is assumed to be 0.5 s for each tangent height for single-scan data, and is assumed to be 15 s for data averaged over 30 scans, which corresponds to a (day-time) half-daily zonal mean for mid-latitude regions. The limb scan step is 2 km for tangent altitudes from 8 to 60 km. The retrieval height grid is set to be 2 km and 4 km for the standard products and the research products, respectively, in the 8-60 km region.

The results are shown in the figures shown below. The single scan profiles are shown for the standard products, and the 30-scan averaged profiles, which consist of the 5° -averaged zonal mean profiles, are shown for the research products.

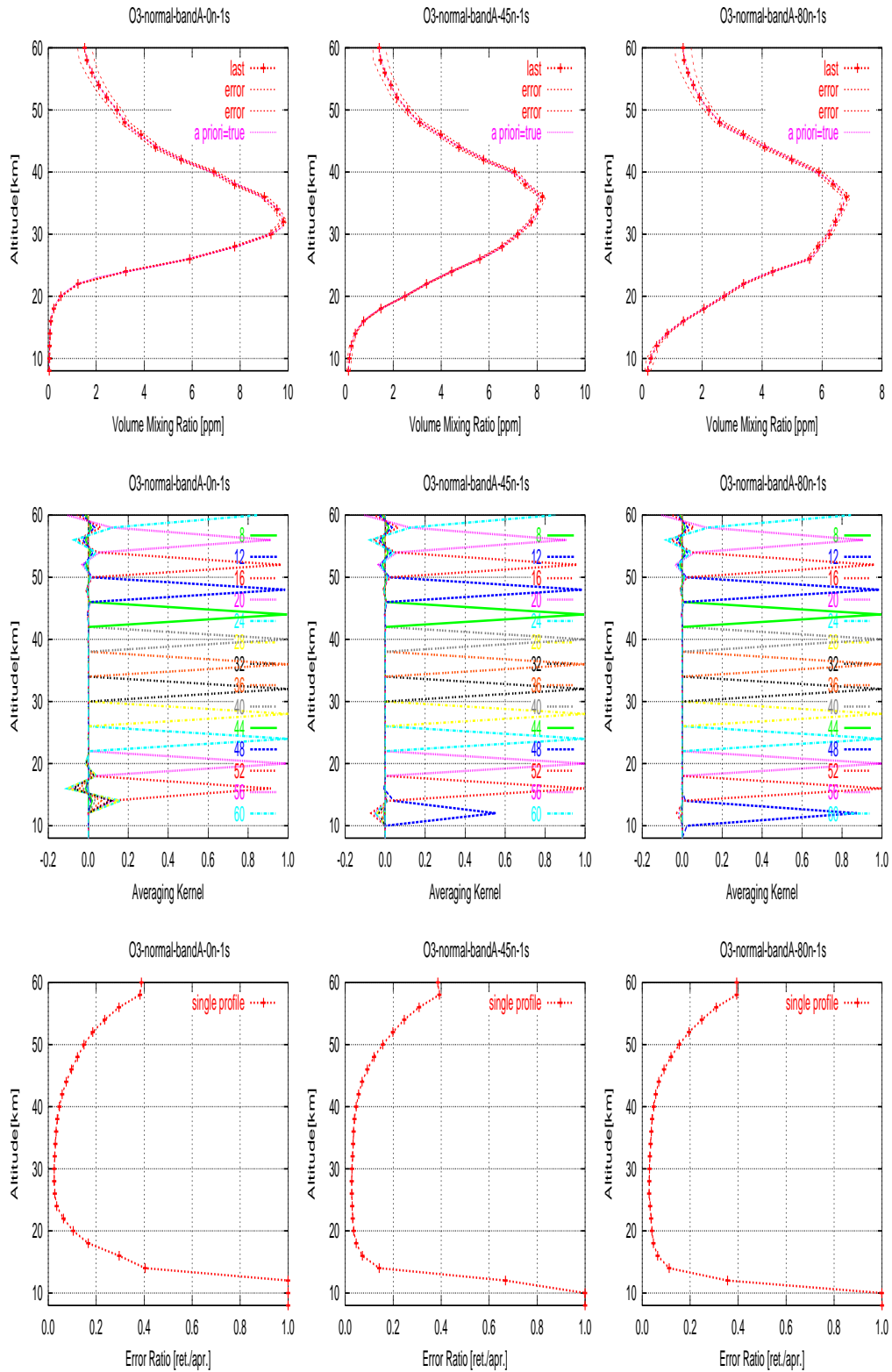


Figure A.1 Retrieval simulation results for single-scan data in BAND-A for standard profiles of normal O₃ at latitudes of 0°N (*left*), 45°N (*center*), and 80°N (*right*). *Top*: Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are assumed to coincide with true profiles; *Middle* Averaging kernels; *Bottom*: Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 50 % of the *a priori* volume mixing ratios.

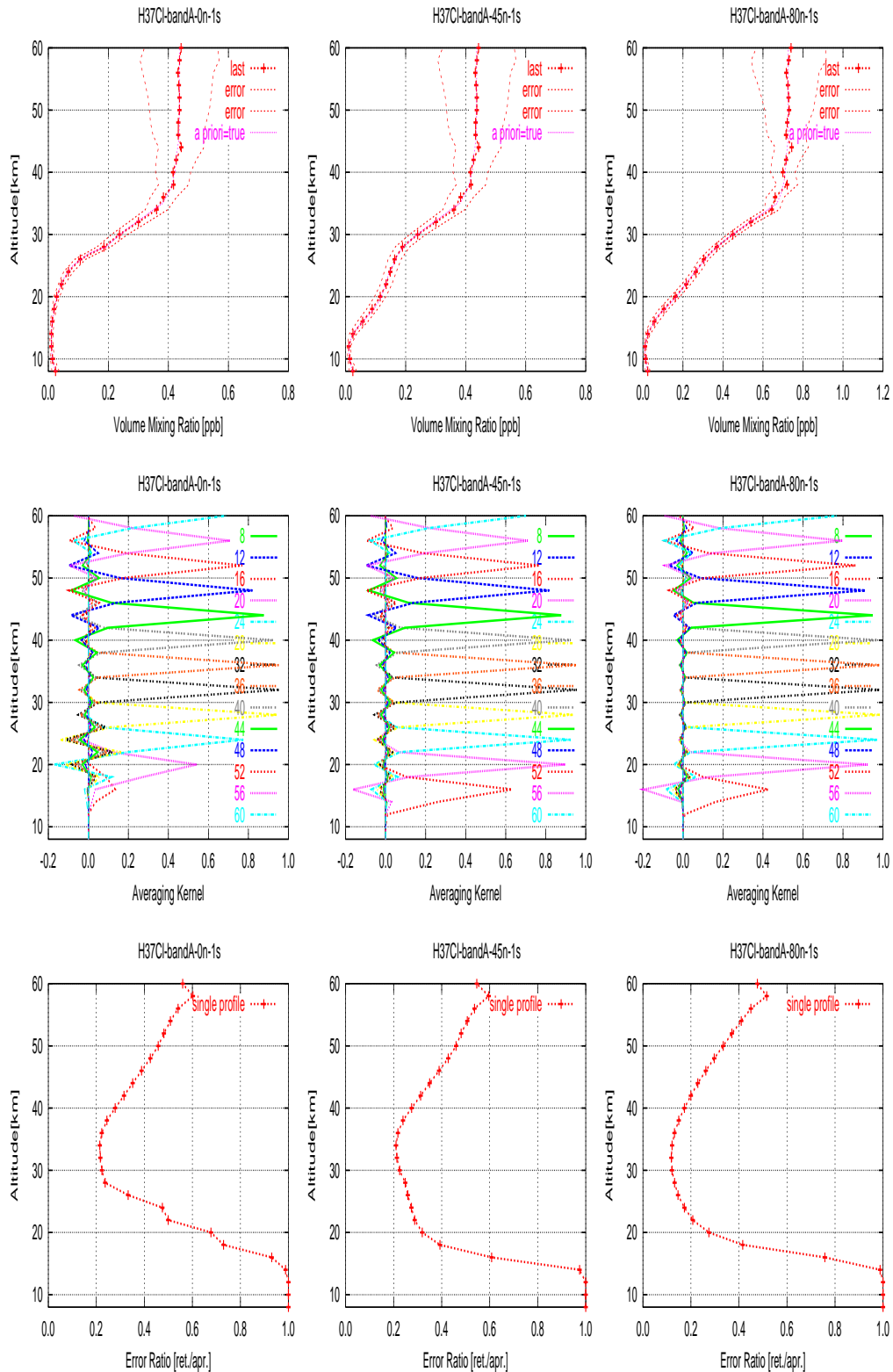


Figure A.2 Retrieval simulation results for single-scan data in BAND-A for standard profiles of H^{37}Cl at latitudes of 0°N (left), 45°N (center), and 80°N (right). *Top*: Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are assumed to coincide with true profiles; *Middle* Averaging kernels; *Bottom*: Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 50 % of the *a priori* volume mixing ratios.

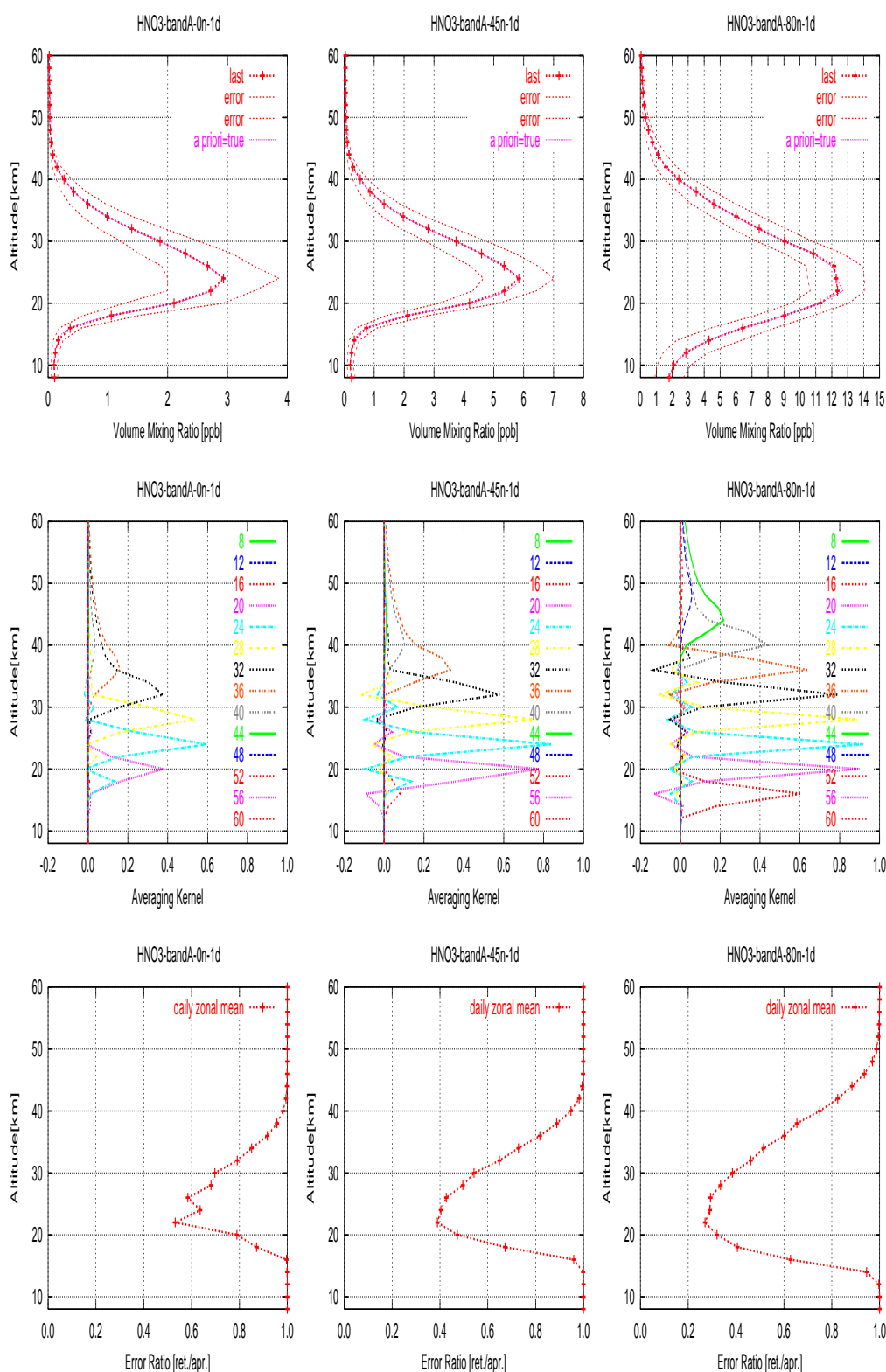


Figure A.3 Retrieval simulation results for 30-scan averaged data in BAND-A for standard profiles of HNO_3 at latitudes of 0°N (*left*), 45°N (*center*), and 80°N (*right*). *Top*: Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are assumed to coincide with true profiles; *Middle* Averaging kernels; *Bottom*: Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 50 % of the *a priori* volume mixing ratios.

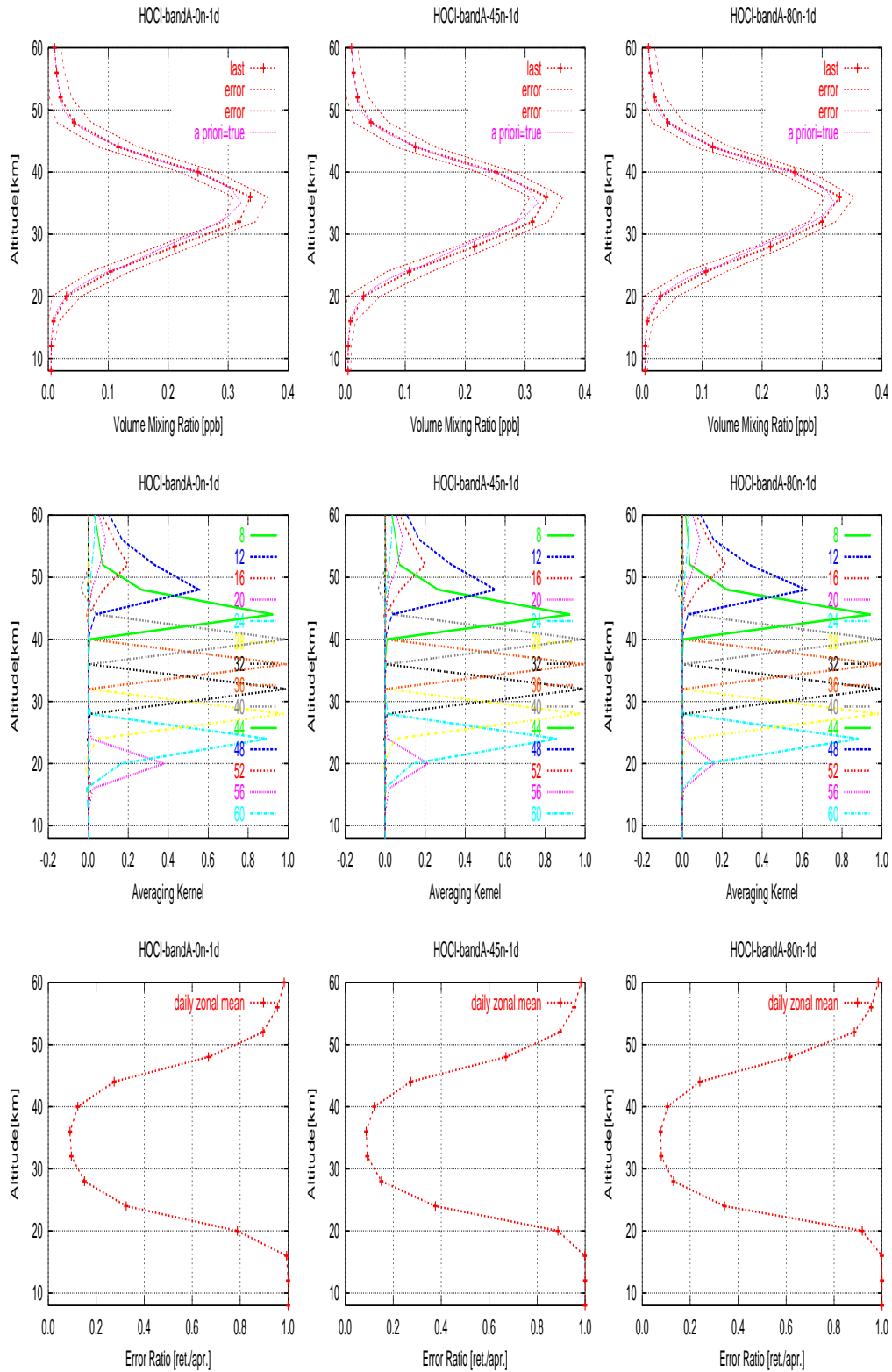


Figure A.4 Retrieval simulation results for 30-scan averaged data in BAND-A for standard profiles of HOCl at latitudes of 0°N (*left*), 45°N (*center*), and 80°N (*right*). *Top*: Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are assumed to coincide with true profiles; *Middle* Averaging kernels; *Bottom*: Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 100 % of the *a priori* volume mixing ratios.

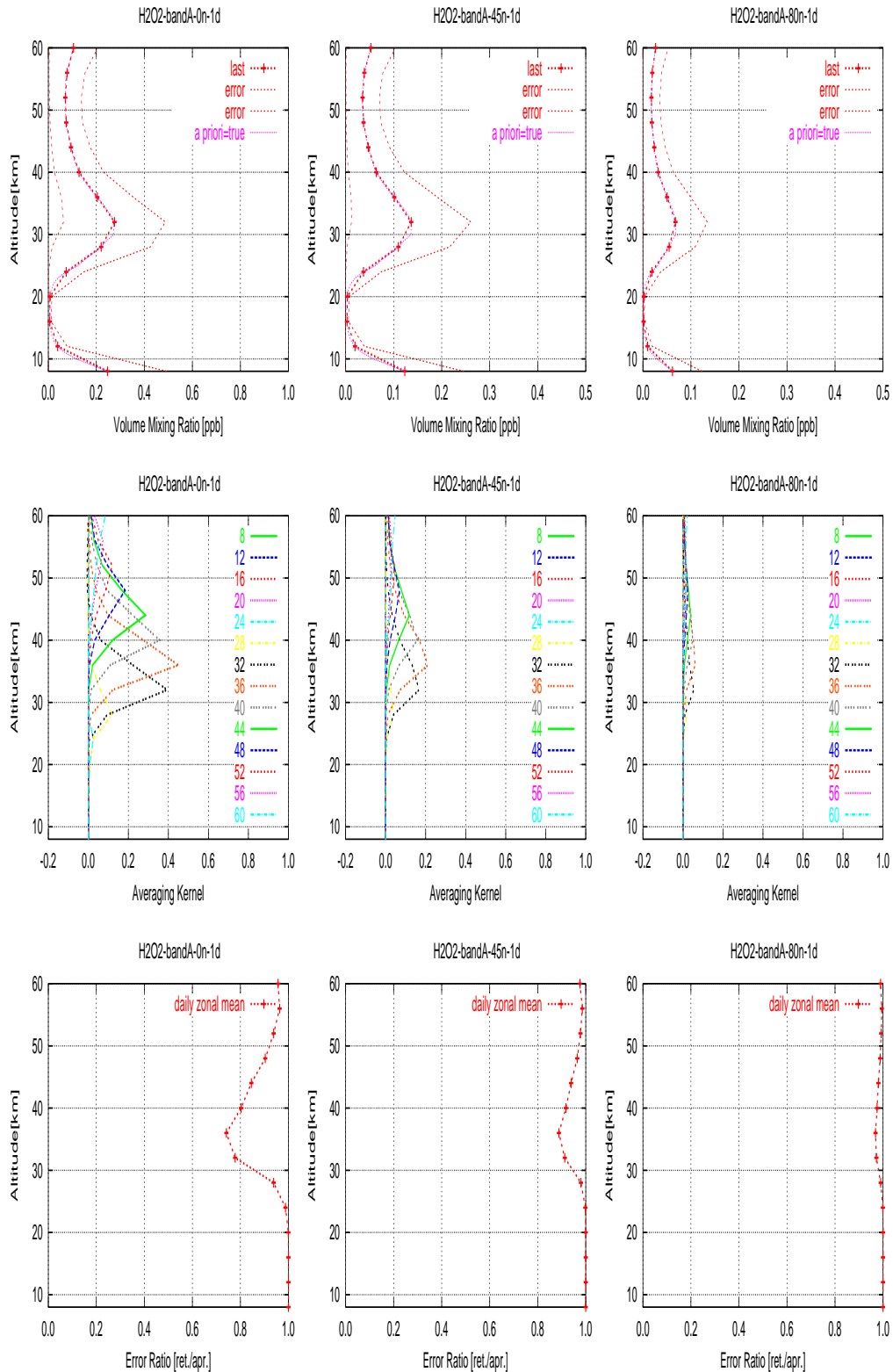


Figure A.5 Retrieval simulation results for 30-scan averaged data in BAND-A for standard profiles of H_2O_2 at latitudes of 0°N (left), 45°N (center), and 80°N (right). *Top:* Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are assumed to coincide with true profiles; *Middle* Averaging kernels; *Bottom:* Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 100 % of the *a priori* volume mixing ratios.

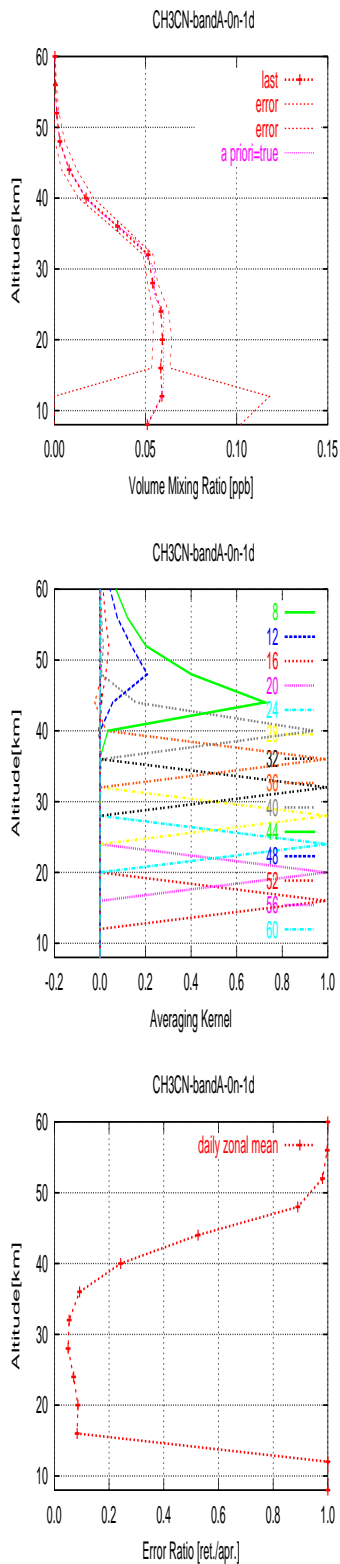


Figure A.6 Retrieval simulation results for 30-scan averaged data in BAND-A for the standard profile of CH_3CN at a latitude of 0°N . *Top:* Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are assumed to coincide with true profiles; *Middle:* Averaging kernels; *Bottom:* Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 100 % of the *a priori* volume mixing ratios.

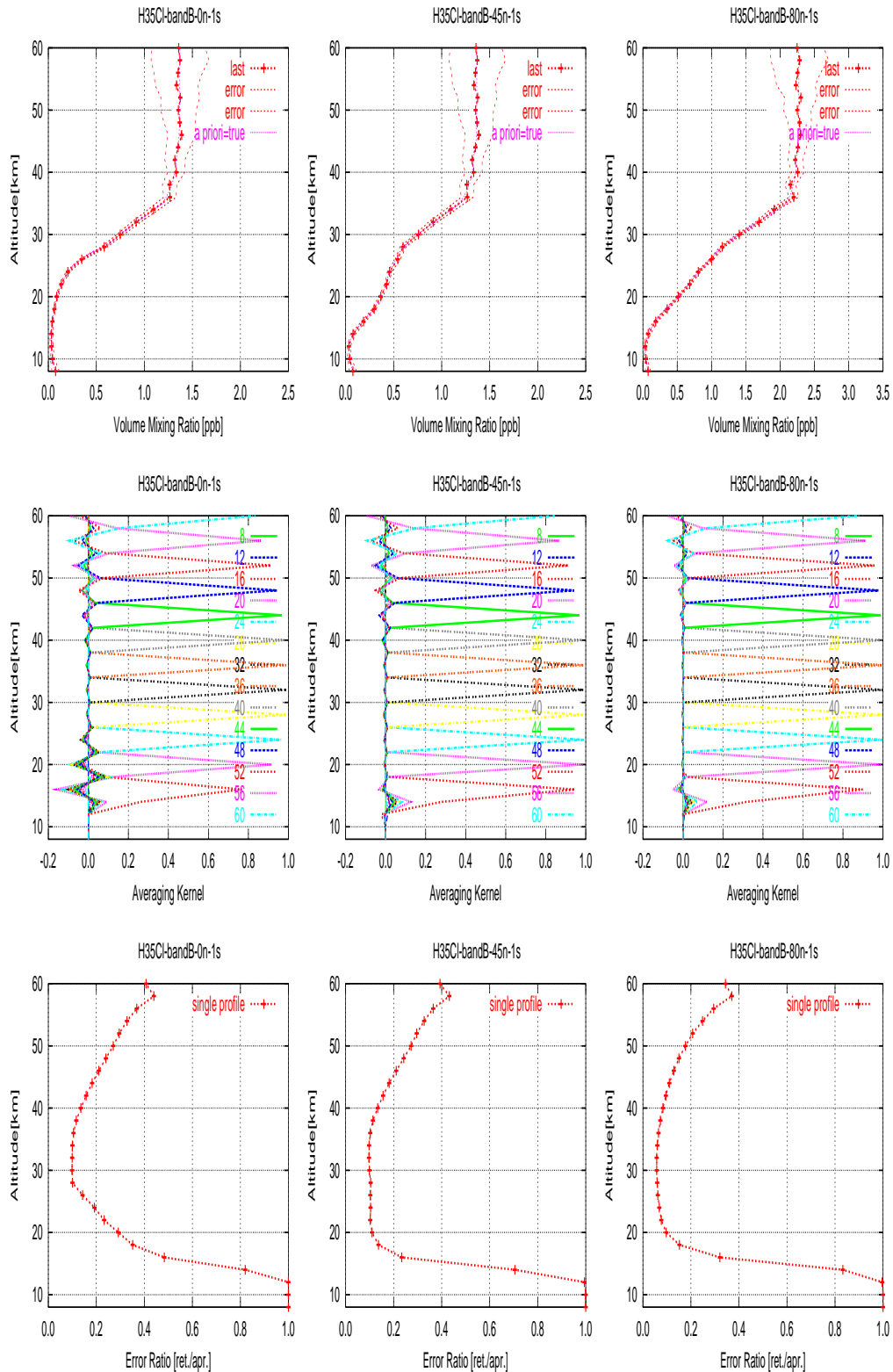


Figure A.7 Retrieval simulation results for single-scan data in BAND-B for standard profiles of $H^{35}Cl$ at latitudes of $0^\circ N$ (left), $45^\circ N$ (center), and $80^\circ N$ (right). *Top*: Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are assumed to coincide with true profiles; *Middle* Averaging kernels; *Bottom*: Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 50 % of the *a priori* volume mixing ratios.

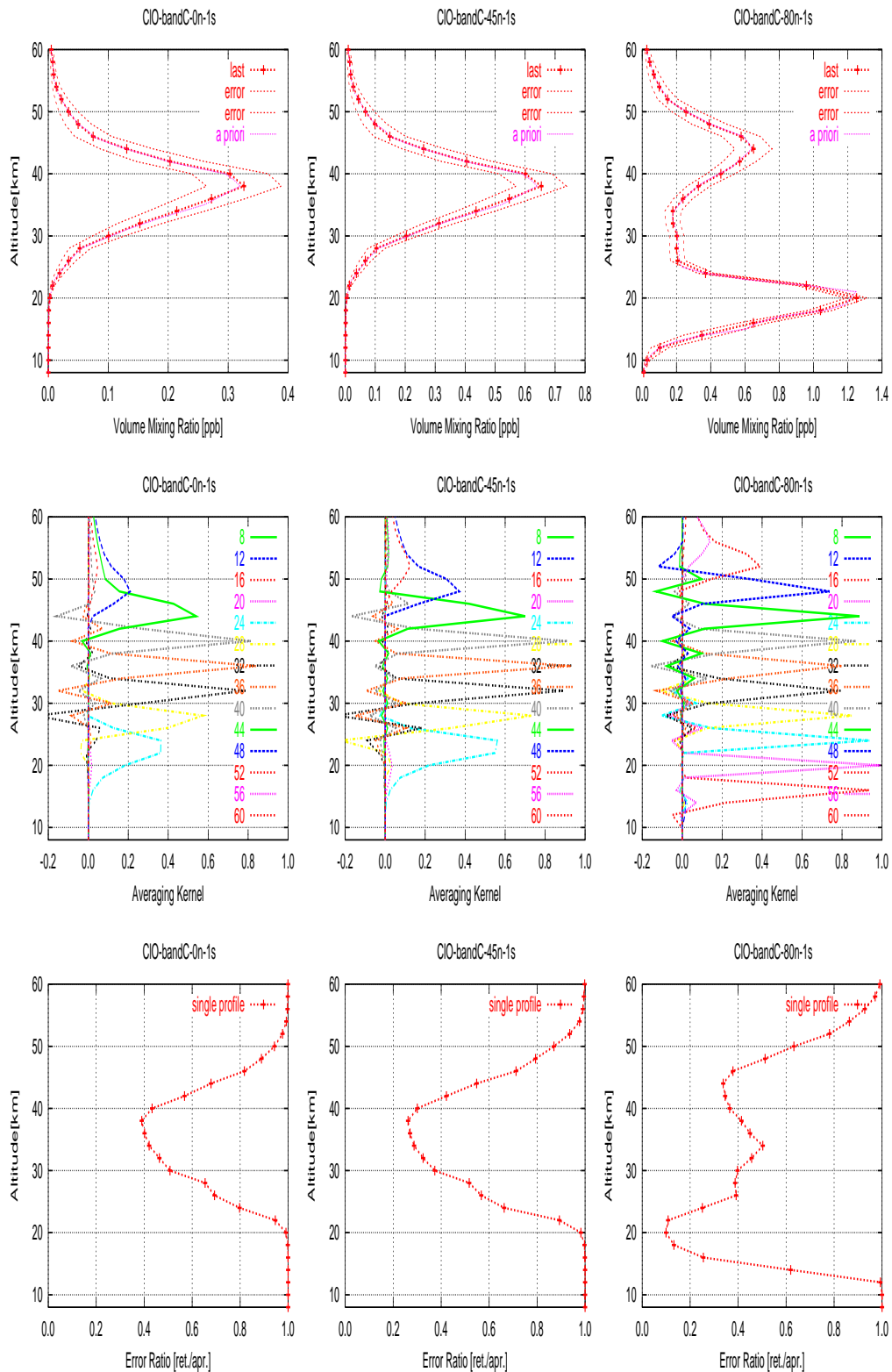


Figure A.8 Retrieval simulation results for single-scan data in BAND-C for standard profiles of ClO at latitudes of 0°N (*left*), 45°N (*center*), and 80°N (*right*). *Top*: Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are assumed to coincide with true profiles; *Middle* Averaging kernels; *Bottom*: Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 50 % of the *a priori* volume mixing ratios.

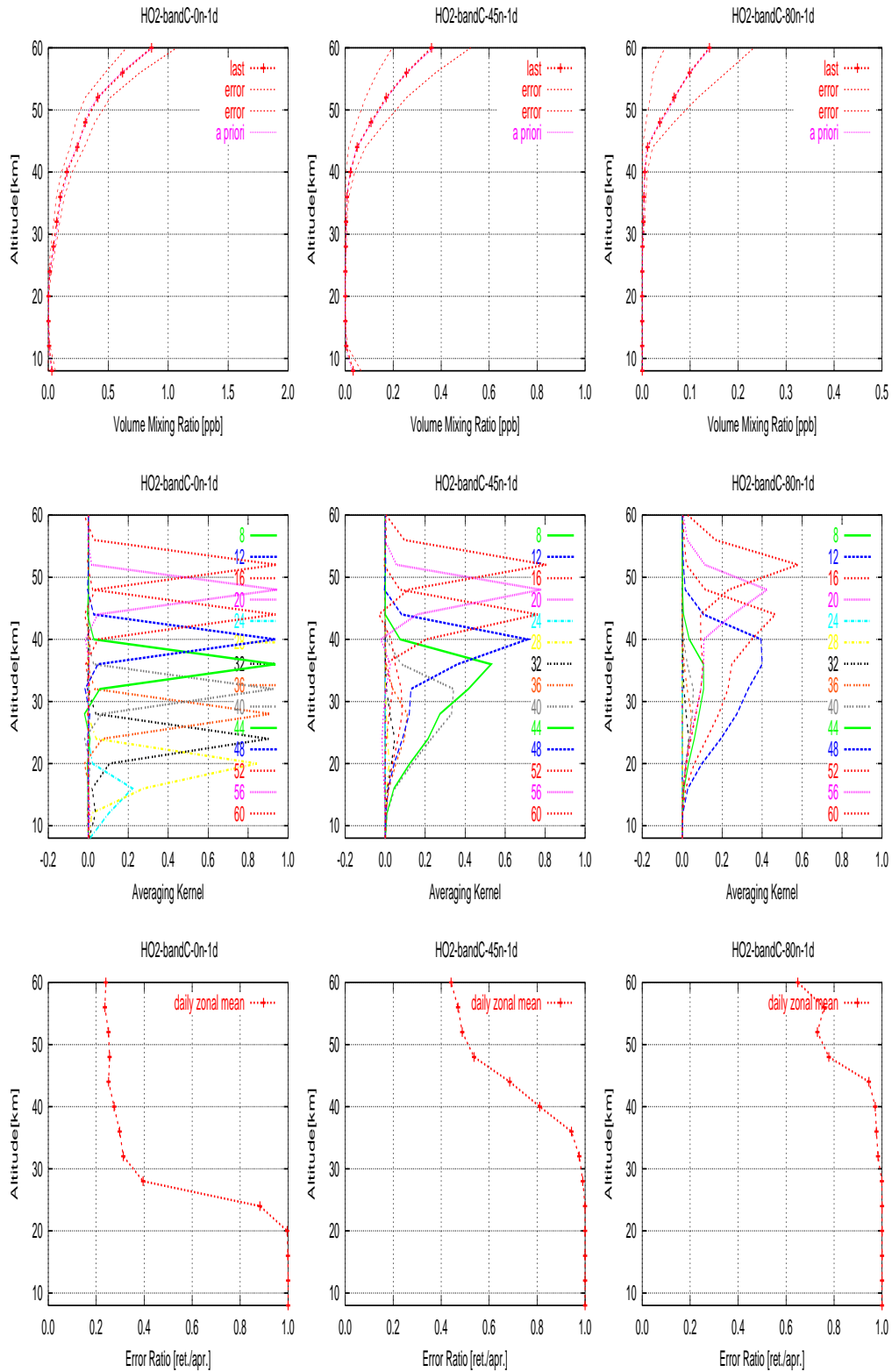


Figure A.9 Retrieval simulation results for 30-scan averaged data in BAND-C for standard profiles of HO₂ at latitudes of 0°N (*left*), 45°N (*center*), and 80°N (*right*). *Top*: Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are assumed to coincide with true profiles; *Middle* Averaging kernels; *Bottom*: Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 100 % of the *a priori* volume mixing ratios.

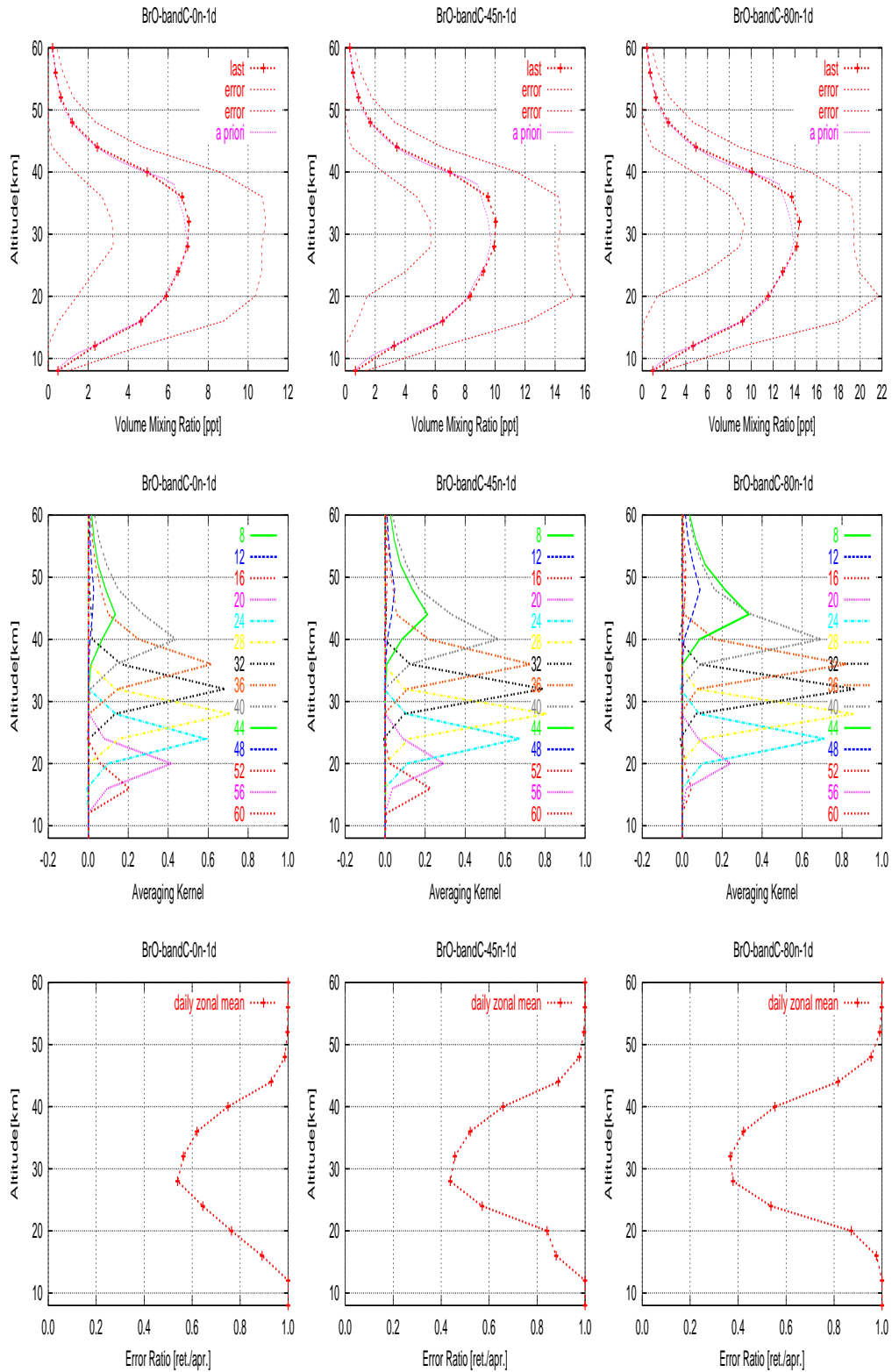


Figure A.10 Retrieval simulation results for 30-scan averaged data in BAND-C for standard profiles of H^{35}Cl at latitudes of 0°N (left), 45°N (center), and 80°N (right). *Top*: Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are assumed to coincide with true profiles; *Middle* Averaging kernels; *Bottom*: Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 100 % of the *a priori* volume mixing ratios.

A.2 Isotope Observations

Simulation setup is the same as that for the radical observations for forward model calculations.

The *a priori* profiles of ozone isotopes are assumed those derived for the standard mid-latitude profile of ozone by applying the atomic-oxygen isotope abundances estimated from the SMOW (Standard Mean Ocean Water) isotope ratio. The true profiles are assumed to be enriched profiles in the simulation. The 11-percent enriched profiles are given at 8-60 km. The *a priori* standard deviations are assumed to be 10 percent of the corresponding *a priori* profiles. The measurement error covariance matrix \mathbf{S}_e is given from the measurement noise, $\sigma = (T_{sys} + T_{atm})/\sqrt{B\tau}$, where T_{sys} is the receiver system noise temperature assumed to be 500 K, and T_{atm} is the antenna temperature, B is the equivalent noise bandwidth assumed to be 0.8 MHz, τ is the integration time. The limb scan step is 2 km for tangent altitudes from 8 to 60 km, and the retrieval height grid and the weighting function step are 5 km. The averaging kernels are plotted every 5 km.

From initial retrieval simulations, it is found that for the here investigated 4 ozone isotopes excluding normal ozone, single-scan spectra of nominal integration time 0.5 s per tangent height have to be averaged over 30 spectra, which correspond to half-daily (day time) zonal means at mid-latitudes from the SMILES observation.

There are 73 transitions of the ozone isotopes in the SMILES observation frequency bands. As relatively isolated spectra of asymmetric-17 O₃ (¹⁷OOO) and asymmetric-18 O₃ (¹⁸OOO), and symmetric-17 O₃ (O¹⁷OO) are found in the Band-C, this upper sideband is therefore better to observe ozone isotopes. The spectral intensity at each transition frequency of the asymmetric ozone isotopes is less than 20 K both substituted of ¹⁸O and ¹⁷O at the tangent height range 10- 60 km at Band-C. Symmetric-17 O₃, has transitions in all three Bands. As symmetric-18 O₃ (O¹⁸OO) has three transitions with very weak intensity in Band-A and B, it should be difficult to observe and to obtain the enrichment in the daily zonal means. If the monthly mean profiles are obtained with stable instrumental conditions, the discussion of the symmetric-18 O₃ height profiles will be possible.

The retrievals were performed for the 30-scan averaged data, which correspond to half-daily (12 hour day time) zonal means. The retrieval results and averaging kernels, absolute error, and relative error are shown for 4 ozone isotopes including normal ozone.

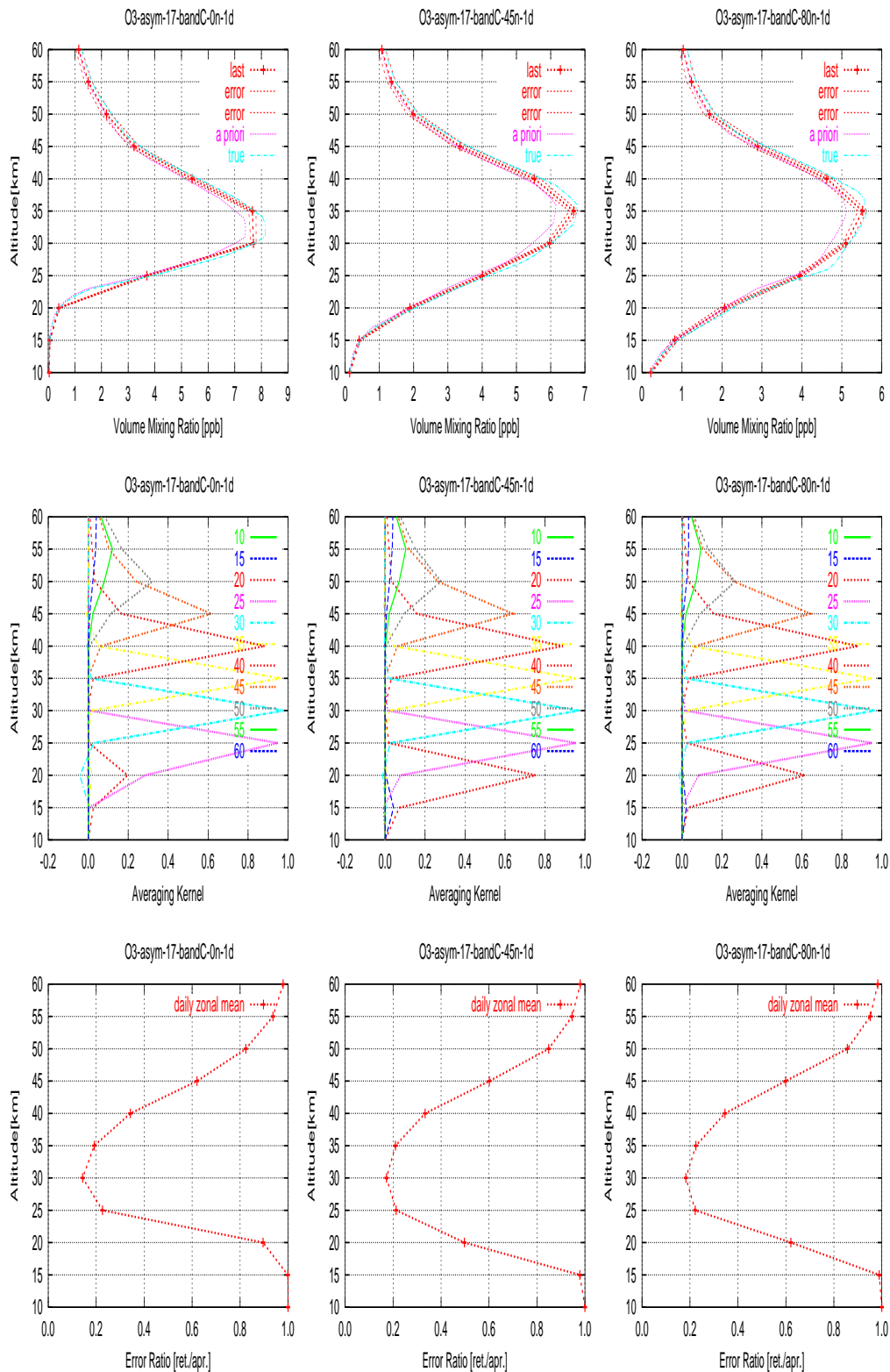


Figure A.11 Retrieval simulation results for 30-scan averaged data in BAND-C for asymmetric-17 O₃ at latitudes of 0°N (*left*), 45°N (*center*), and 80°N (*right*). *Top:* Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are here assumed to be 90% of true profiles; *Middle* Averaging kernels; *Bottom:* Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 10 % of the *a priori* volume mixing ratios.

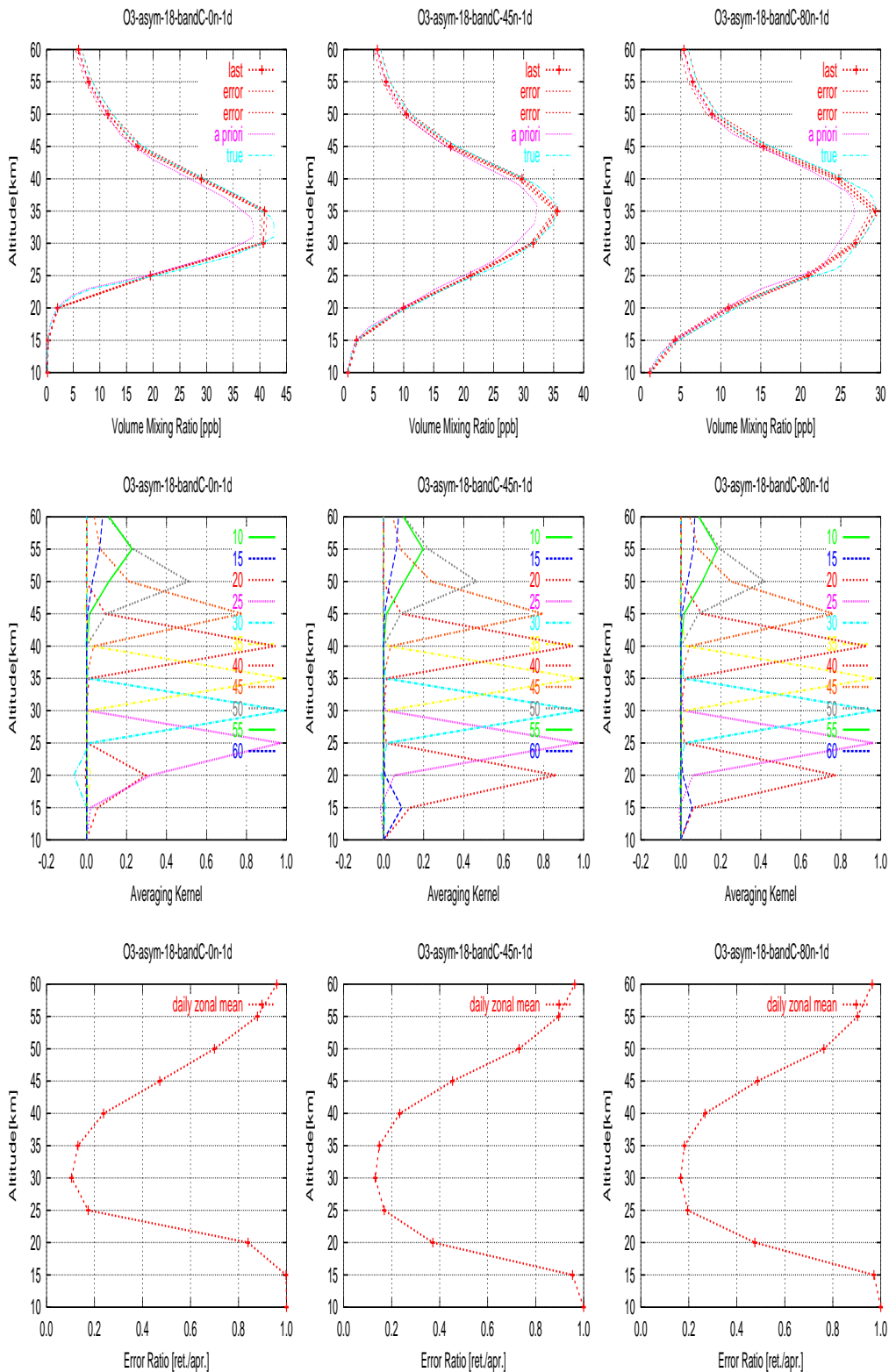


Figure A.12 Retrieval simulation results for 30-scan averaged data in BAND-C for asymmetric-18 ozone at latitudes of 0°N (*left*), 45°N (*center*), and 80°N (*right*). *Top*: Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are here assumed to be 90% of the true profile; *Middle* Averaging kernels; *Bottom*: Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 10 % of the *a priori* volume mixing ratios.

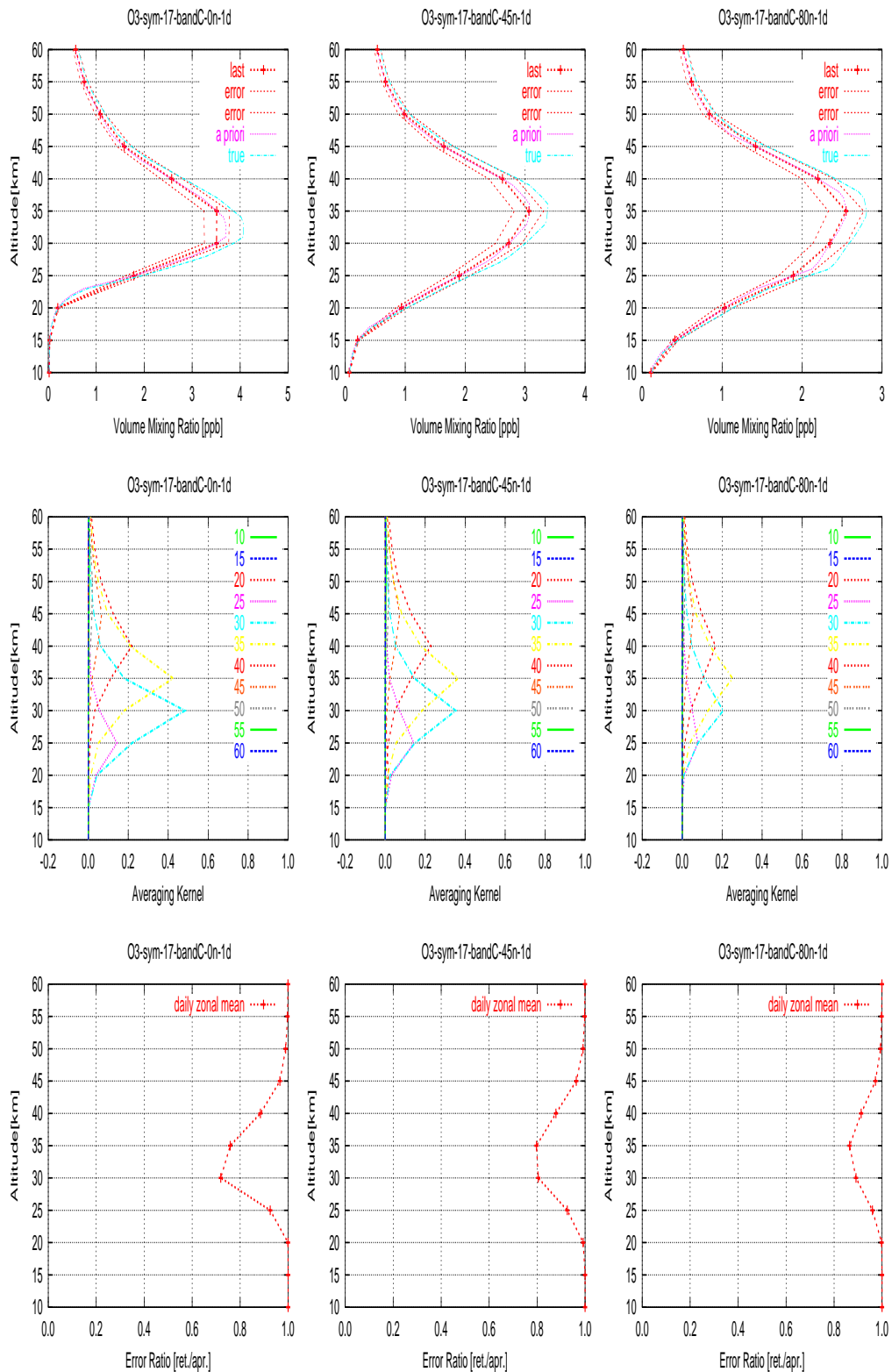


Figure A.13 Retrieval simulation results for 30-scan averaged data in BAND-C for symmetric-17 ozone at latitudes of 0°N (*left*), 45°N (*center*), and 80°N (*right*). *Top*: Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are here assumed to be 90% of the true profiles; *Middle* Averaging kernels; *Bottom*: Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 10 % of the *a priori* volume mixing ratios.

A.3 Mesospheric Observation

We performed the error estimation for ozone, HCl, and HO₂ in the mesosphere, using the model atmosphere for the polar, mid-latitude, and equatorial regions, using the same conditions as those used for the radical analysis except for weighting function height step and observation height step. The errors for each 10 km partial column is obtained.

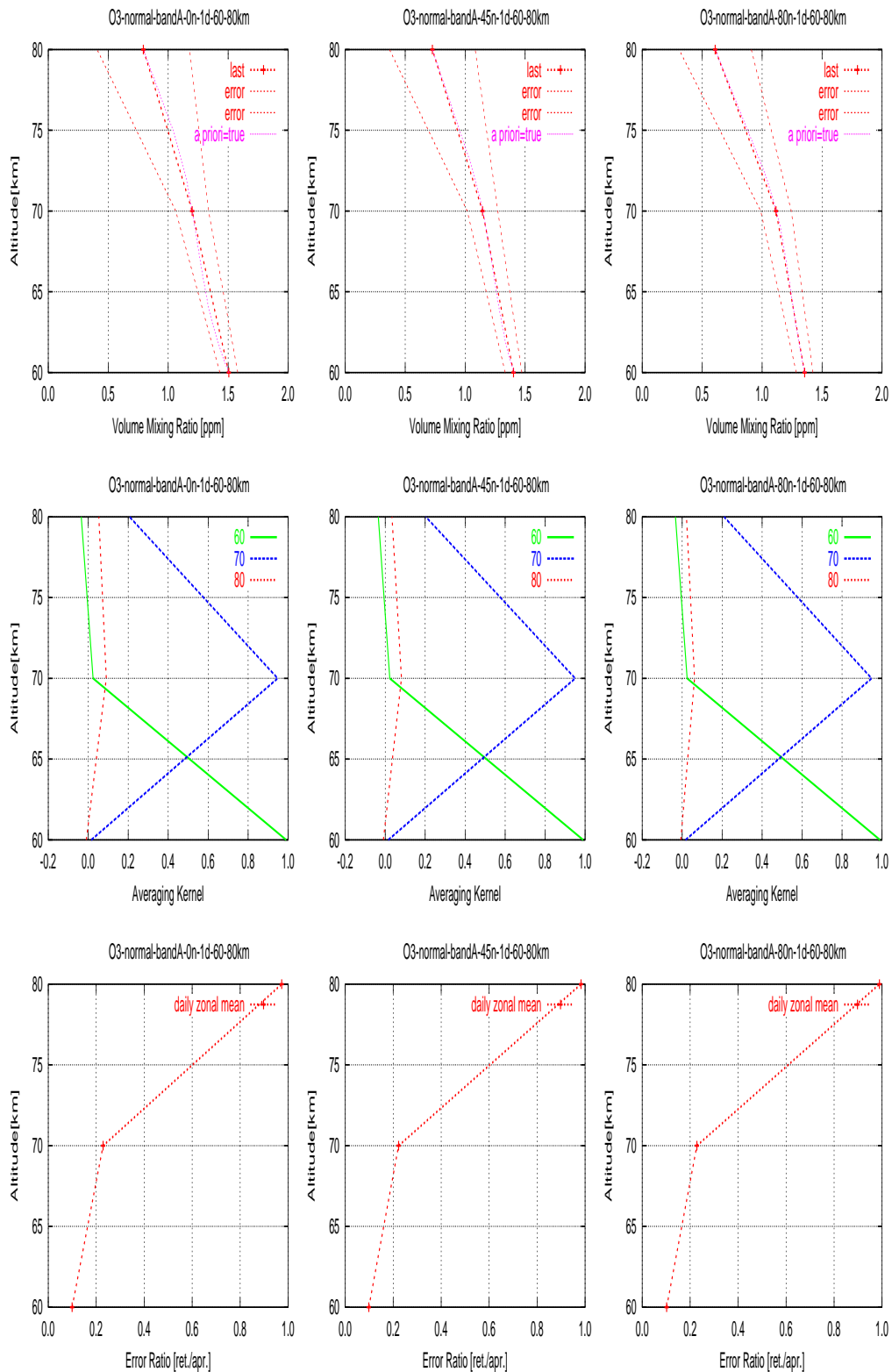


Figure A.14 Retrieval simulation results for 30-scan averaged data in BAND-A for the standard profiles of normal O₃ in the mesosphere at latitudes of 0°N (*left*), 45°N (*center*), and 80°N (*right*). *Top*: Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are assumed to coincide with true profiles; *Middle* Averaging kernels; *Bottom*: Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 50 % of the *a priori* volume mixing ratios.

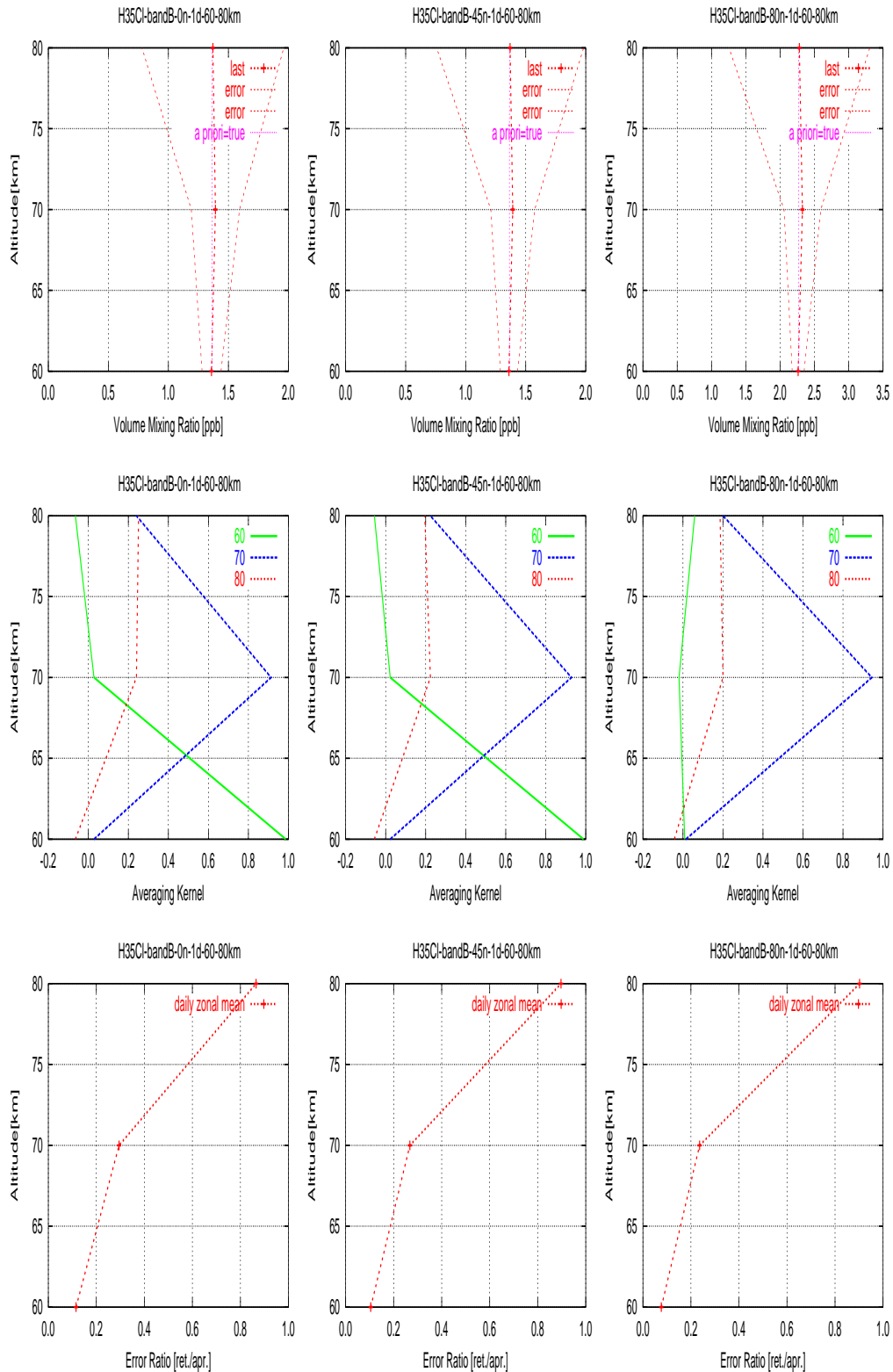


Figure A.15 Retrieval simulation results for 30-scan averaged data in BAND-B for the standard profiles of H^{35}Cl in the mesosphere at latitudes of 0°N (left), 45°N (center), and 80°N (right). *Top:* Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are assumed to coincide with true profiles; *Middle:* Averaging kernels; *Bottom:* Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 50 % of the *a priori* volume mixing ratios.

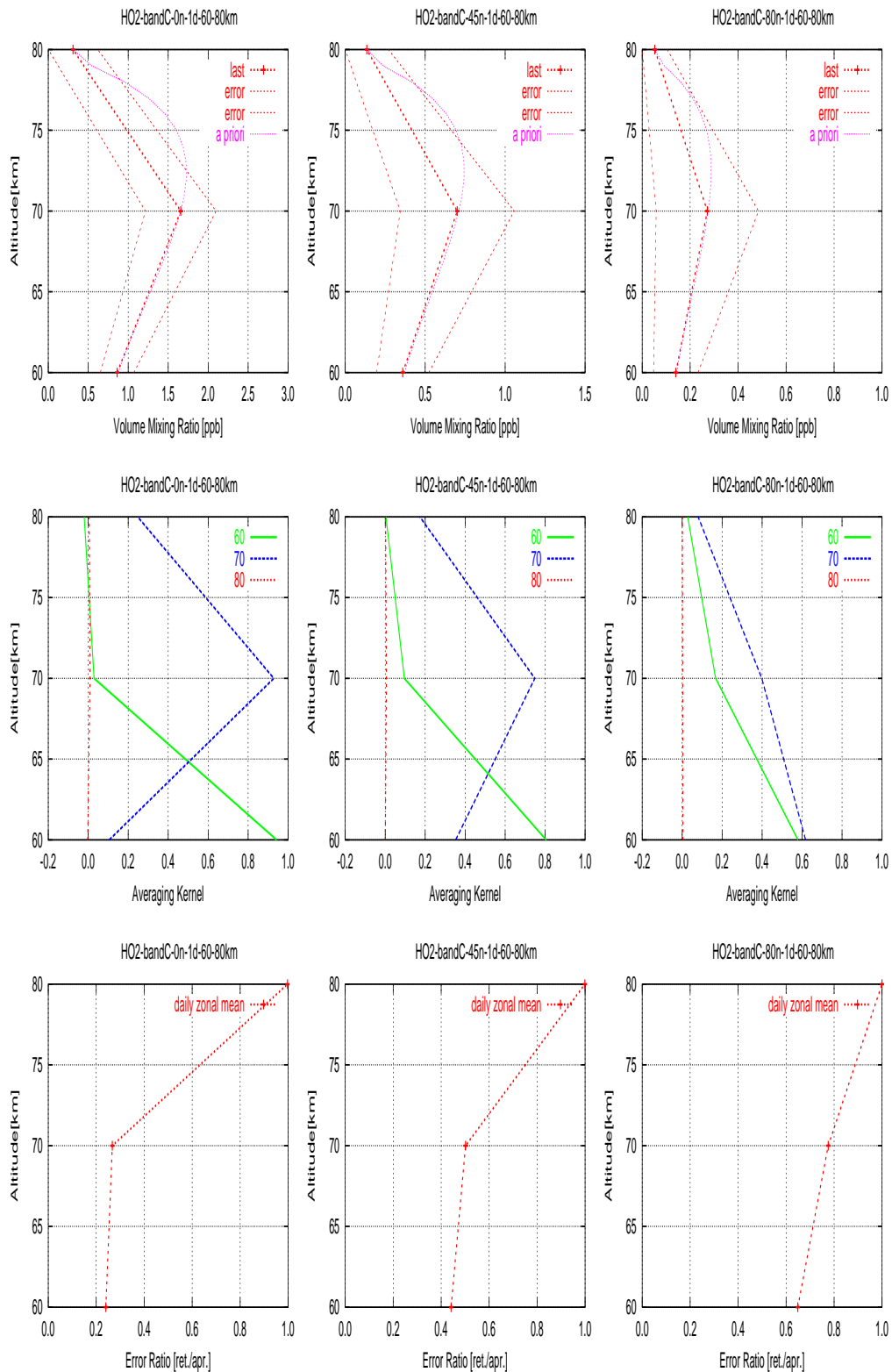


Figure A.16 Retrieval simulation results for 30-scan averaged data in BAND-C for the standard profiles of HO₂ in the mesosphere at latitudes of 0°N (*left*), 45°N (*center*), and 80°N (*right*). *Top*: Retrieved volume mixing ratios are shown with *rms* retrieval error bounds and *a priori* profiles which are assumed to coincide with true profiles; *Middle* Averaging kernels; *Bottom*: Retrieval error ratios defined as the ratio of the retrieval errors to the *a priori* errors which are assumed to be 100 % of the *a priori* volume mixing ratios.