Supporting Information

Vertical profile observations of water vapor deuterium excess in the lower troposphere

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Supporting Figures

**Figure S1.** Comparison of Rayleigh distillation curves calculated using a single fractionation factor ($\alpha$) defined by the temperature at the lifting condensation level (LCL) and temperature-varying $\alpha$ values. The Rayleigh curve comparison is done for the second vertical profile (VP2) conducted on the stratocumulus cloud (STC) case study.

**Figure S2.** Rapid Refresh (RAP) Model ambient temperature and dewpoint profiles for the Indianapolis International Airport (KIND) from 22:00 EDT on 17 March 2018 (Research Flight 08; RF08) and 11:00 - 17:00 EDT on 18 March 2016 (developing boundary layer case study; DBL) showing incorporation of the 17 March 2016 residual layer into the DBL flight day’s boundary layer.

**Figure S3*.** Progression of satellite cloud cover images of the Indianapolis area on 4 March 2016, the stratocumulus cloud (STC) case study day, from 11:45 – 17:45 local time (EDT). The GIF shows that the stratocumulus cloud layer moves eastward and thins over the course of the afternoon.

**Figure S4*.** Progression of satellite cloud cover images of the Indianapolis area on 18 March 2016, the developing boundary layer (DBL) case study day, from 13:45 – 17:30 EDT. The GIF shows that a cloud layer develops and moves into the study area at the end of the flight. Moistening of the mid-troposphere on DBL is also shown in Fig. S2 and Fig. S5.

**Figure S5.** Free troposphere (FT) measurements of $\delta^{18}$O, $\delta$D, d-excess.

**Figure S6.** Weather map of mid-troposphere (3-5.5 km) relative humidity on the developing boundary layer (DBL; 18 March 2016) case study at 17:00 EDT.

*Figure S3 and S4 are presented separately as Graphics Interchange Format (GIF) files.*
Figure S1. Comparison of Rayleigh distillation curves calculated using a single fractionation factor ($\alpha$) defined by the temperature at the lifting condensation level (LCL) and temperature-varying $\alpha$ values. The Rayleigh curve comparison is shown for temperatures measured along the second vertical profile (VP2) conducted on the stratocumulus cloud (STC) case study.
Figure S2. Rapid Refresh (RAP) Model ambient temperature and dewpoint profiles for the Indianapolis International Airport (KIND) from 22:00 EDT on 17 March 2018 (Research Flight 08; RF08) and 11:00 - 17:00 EDT on 18 March 2016 (Developing Boundary Layer case study; DBL). The progression of figures shows a near dry adiabatic lapse rate (DALR; white line) persisted from 17 March 2018 into the afternoon of 18 March 2018. With time, the 17 March 2018 residual layer (with nearly a DALR) is incorporated into the 18 March 2018 boundary layer.
Figure S3 and Figure S4 are Graphics Interchange Format files (GIFs) that show cloud cover evolution in the Indianapolis area on 4 March 2016, the stratocumulus cloud (STC) case study day, and 18 March 2016, the developing boundary layer (DBL) case study day. The files can be downloaded from the Supporting Information page.

Figure S3*. Progression of satellite cloud cover images of the Indianapolis area on 4 March 2016, the stratocumulus cloud (STC) case study day, from 11:45 – 17:45 local time (EDT). The GIF shows that the stratocumulus cloud layer moves eastward and thins over the course of the afternoon.

Figure S4*. Progression of satellite cloud cover images of the Indianapolis area on 18 March 2016, the developing boundary layer (DBL) case study day, from 13:45 – 17:30 EDT. The GIF shows that a cloud layer develops and moves into the study area at the end of the flight. Moistening of the mid-troposphere on DBL is also shown in Fig. S2 and Fig. S5.
Figure S5: Free troposphere (FT) measurements of (a) $\delta^{18}$O, (b) $\delta$D, (c) d-excess below 4000 ppmv H$_2$O$_v$ during the research flights in Indianapolis and Washington, D.C.-Baltimore. The dashed line in (c) corresponds to d-excess = 10 ‰, the global average of precipitation, for reference.
Figure S6. Weather map of mid-troposphere (3-5.5 km) relative humidity on the Developing Boundary Layer (DBL; 18 March 2016) case study day at 17:00 local time (EDT). Figure S6 shows moistening ahead of a shortwave trough. Indiana is outlined by an orange box, and Indianapolis is indicated with a star. The green areas show relative humidity greater than 80%. This weather map supports observations of elevated H$_2$O mole fraction in the free troposphere during the fourth vertical profile (VP4) on DBL (Fig. 6d).