SUPPLEMENTARY INFORMATION

Shortwave Radiative Impacts of the Asian Tropopause Aerosol Layer (ATAL) using Balloon-borne In-situ measurements at three distinct locations in India

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S1. Identification of clouds using relative humidity thresholds

We used an approach by combining the vertical gradients of air temperature and relative humidity (RH) and the altitude-dependent thresholds of RH to determine the clouds in the lower and free troposphere as described in Xu et al., (2023). For this purpose, the profiles of temperature, pressure, and RH were obtained from the radiosonde soundings. In this approach, we first converted the portion of radiosonde-observed RH (RH_{liq}) to the RH_{ice} (relative humidity with respect to ice) and to RH_{mixed} (relative humidity with respect to mixed phase) using the following equations

$$RH(z) = \begin{cases} RH_{liq},\ T(z) > 0°C \\ RH_{mix}(z),\ -20°C < T(z) < 0°C \\ RH_{ice}(z),\ T(z) \leq 20°C \end{cases} \quad (1)$$

where the RH_{ice} and RH_{mixed} are derived using the saturation vapor pressure in the pure liquid and ice phase with respect to altitude 'z' (e_{liq}(z) and e_{ice} (z) respectively) using the following equations

$$e_{liq}(z) = 6.1078\ exp\left[\frac{17.2693882\ T(z)}{T(z)+237.3}\right] \qquad (2a)$$

$$e_{ice}(z) = 6.1078\ exp\left[\frac{21.8745584\ (T(z)-3)}{T(z)+265.5}\right] \qquad (2b)$$

$$RH_{ice}(z) = RH_{liq}(z) \times \frac{e_{liq}(z)}{e_{ice}(z)} \qquad (2c)$$

where temperature (T) and vapour pressures are in oC and hPa, respectively. The RH_{mixed} is calculated using the equation,

$$RH_{mixed}(z) = \frac{-20-T(z)}{-20}\ RH_{liq}(z) + \frac{T(z)}{-20} RH_{ice}(z) \qquad (2d)$$

After pre-processing the RH(z), by examining the first and second derivatives of RH(z) and T(z) starting from the surface upward, the bases of the moist layers can be detected when the following criteria are satisfied:

$$base: \begin{cases} RH'(z) > 0\ and\ RH'' < 0 \\ T'(z) < 0\ and\ T''(z) > 0 \end{cases} \qquad (3a)$$

$$top: \begin{cases} RH'(z) < 0\ and\ RH'' < 0 \\ T'(z) > 0\ and\ T''(z) > 0 \end{cases} \qquad (3b)$$

The moist layers identified using this method are attributed as cloud layers if the following threshold conditions are satisfied. (1) The base of the moist layer exceeds 280 meters; (2) The thickness of the moist layer is greater than 30.5 meters and 61 meters for bases of less than 2 km and greater than 2 km, respectively; (3) The minimum relative humidity (min-RH) within the moist layer surpasses the corresponding min-RH threshold at the base of the moist layer (Table S1); (4) The maximum relative humidity (max-RH) within the moist layer exceeds the corresponding max-

RH threshold at the base of the moist layer (**Table S1**). Utilizing the max-RH criterion helps prevent the misidentification of some moist layers as cloud layers. Otherwise, the moist layer is excluded from the analysis. Also, if the distance between two adjacent cloud layers is less than 300 m, or if the minimum relative humidity (min-RH) between the continuous cloud layers exceeds the corresponding minimum RH threshold (inter-RH) between consecutive cloud layers (**Table S1**), then these two cloud layers are combined. An example of this method of cloud screening is shown in **Fig. S2**.

S2. The aerosol types considered for the boundary layer and free tropospheric aerosols

The different aerosol mixed type aerosol models taken from Hess et al. (1998) are used for defining the aerosol types within the boundary layer (WBL) and free troposphere (FT) based on the air mass back trajectory analysis across the study locations. Also, we have identified major sectors in and around the Indian region that influence a given air mass originating or passing through (**Section 4** of the main manuscript). A brief description of the different types is as follows:

i) **Maritime Tropical (MT)** (aerosols with low density of water-soluble substances, lower wind speed, and lower amount of sea salt), which are mainly assigned to the air masses reaching over Gadanki within the boundary layer from the southern sector given the relatively clean local conditions here.;

ii) **Maritime Polluted (MP)** (refers to air masses of oceanic origin that are heavily influenced by the highly varying soot and also of anthropogenic water-soluble particles) assigned for the air masses from the eastern and southern sectors that reach over Hyderabad within the boundary layer as the local emissions in an around the city are highly polluted.;

iii) **Continental Average (CA)** (used to describe continental areas with moderate anthropogenic influence) assigned for the air masses originating/passing through the C sector.

iv) **Continental Polluted (CP)** (for areas highly polluted by man-made activities. The mass density of soot is 2 μg m^{-3}, and the mass density of water-soluble substances is more than double that in continental average aerosol) for describing the air masses in the northern sector

v) **Desert (DST)** to describe aerosol over the arid regions which are assigned to the long-range transported air masses from the north-western sector.

Fig. S3 shows the optical properties (SSA and ASY) of the aerosol types specified above at different relative humidity bins.

Supplementary Tables

Table S1: *The altitude-resolved RH thresholds for the cloud screening of the BSR data below 10 km*

Altitude Range	**Min-RH**	**Max-RH**	**Inter-RH**
0-2 km	84%	94%	82%
2-6 km	80%	92%	78%
6-12 km	78%	88%	70%
>12 km	70%	80%	70%

Supplementary Figures

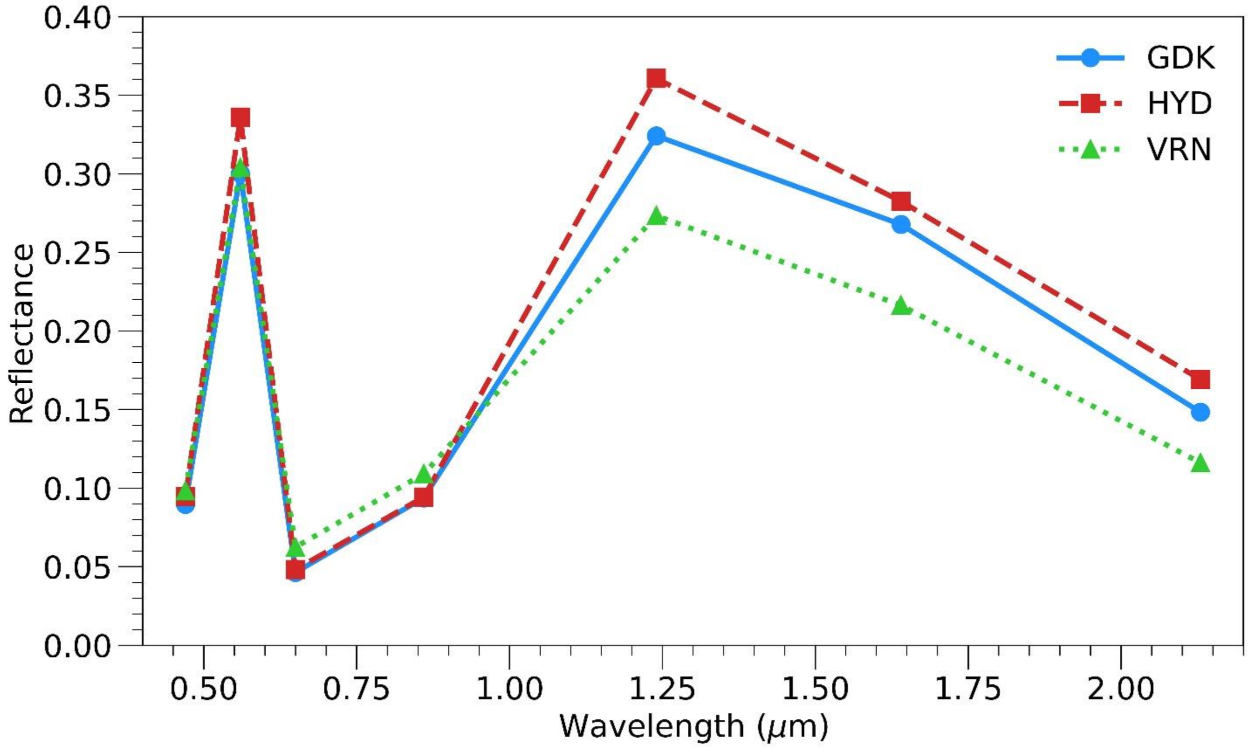

Figure S1: The spectral variation in the surface reflectance across the study locations – Gadanki (GDK), Hyderabad (HYD), and Varanasi (VRN)

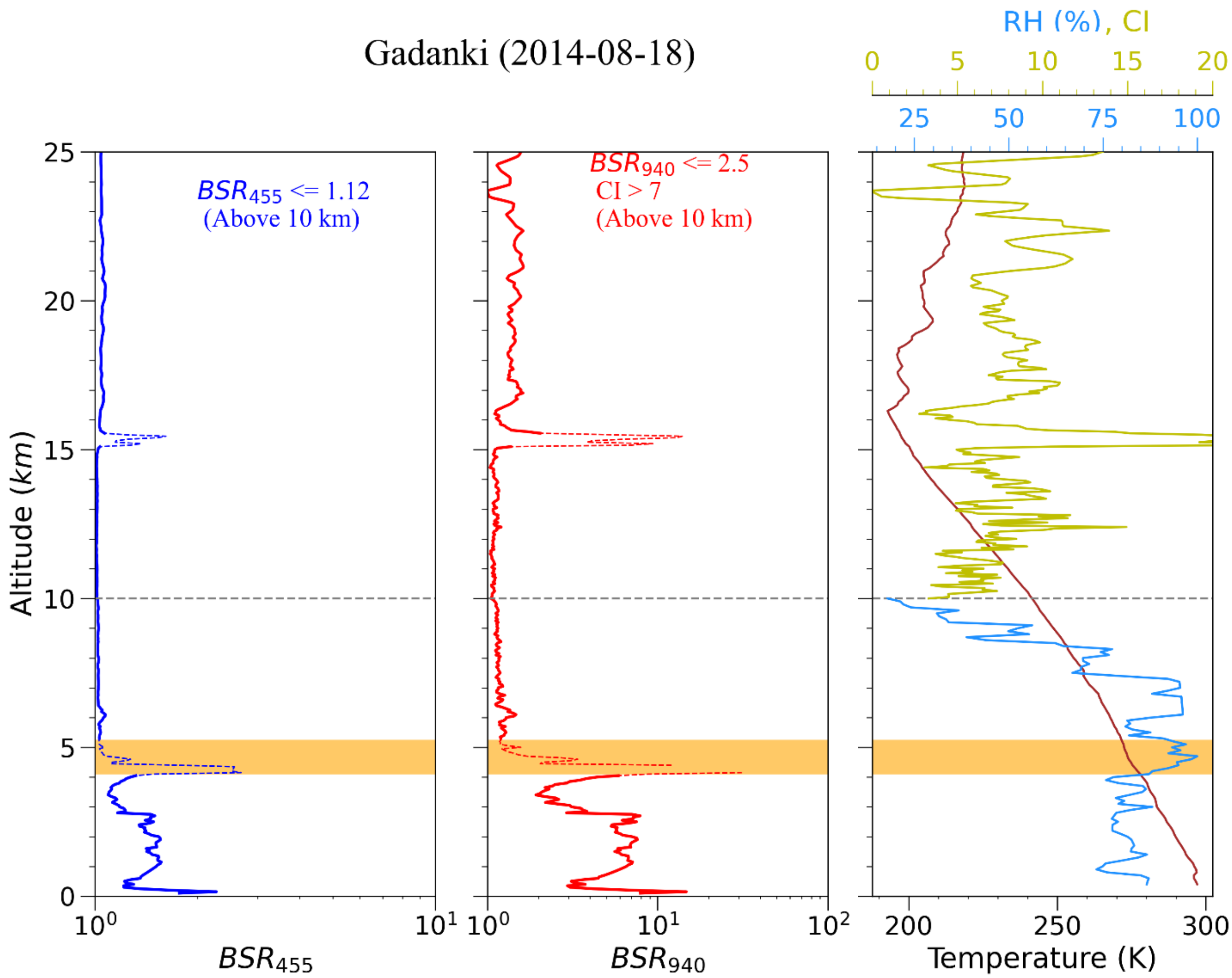

Figure S2*:* The cloud screening procedure for a given COBALD profile is based on the RH thresholds (below 10 km) and thresholds for the backscatter ratio (BSR) and Color Index (CI) (Above 10 km). The shaded region represents the identified cloud. The dashed lines represent the screened-out data.

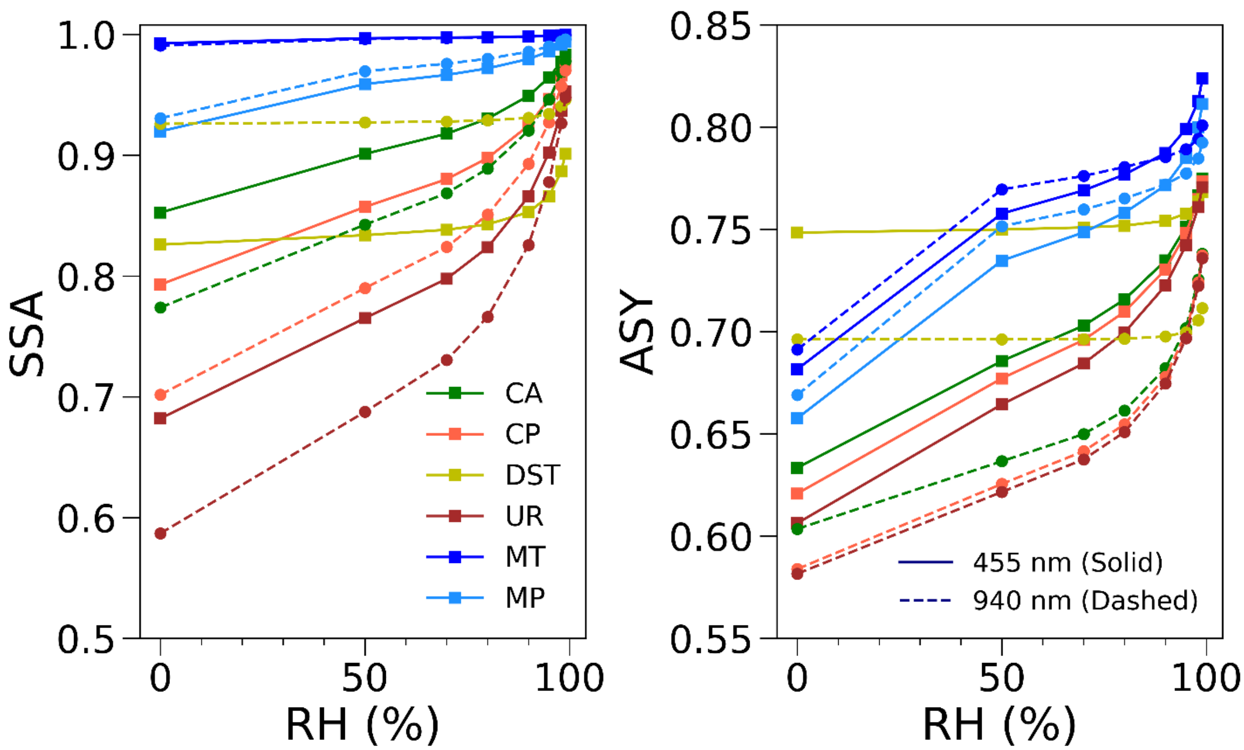

Figure S3: The variation of SSA and ASY for the aerosol types discussed in **Section S2** at different RH bins.

References

Hess, M., Koepke, P., & Schult, I. (1998). Optical Properties of Aerosols and Clouds: The Software Package OPAC. *Bulletin of the American Meteorological Society*, *79*(5), 831–844. https://doi.org/10.1175/1520-0477(1998)079<0831:OPOAAC>2.0.CO;2

Xu, H., Guo, J., Tong, B., Zhang, J., Chen, T., Guo, X., Zhang, J., & Chen, W. (2023). Characterizing the near-global cloud vertical structures over land using high-resolution radiosonde measurements. *Atmospheric Chemistry and Physics*, *23*(23), 15011–15038. https://doi.org/10.5194/acp-23-15011-2023