**S1. Identification and Screening of Cloud Layers in the Lower Atmosphere**

We employed an approach that combined the vertical gradients of air temperature and relative humidity (RH) with altitude-dependent RH thresholds to identify 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 (RHliq) to RHice (relative humidity with respect to ice) and RHmixed (relative humidity with respect to mixed phase) using the following equations:

(1)

where the RHice and RHmixed are derived using the saturation vapor pressure in the pure liquid and ice phase with respect to altitude z (eliq(z) and eice (z), respectively), using the following equations

(2a)

(2b)

(2c)

where temperature (T) and vapour pressures are in °C and hPa, respectively. The RHmixed is calculated using the equation,

(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:

(3a)

(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 in preventing 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 cloud screening method is shown in **Fig. S3**.

**S2. Identification of Aerosol Types in the Lower Atmosphere**

Understanding the aerosol composition in the lower atmosphere is crucial for determining the Single Scattering Albedo (SSA) and Asymmetry Parameter (ASY), which are necessary for radiative transfer calculations across the atmospheric column. For this purpose, we have used the air-mass back-trajectory clustering technique using the HYSPLIT software (Draxler and Hess, 1998). HYSPLIT classifies air trajectories using a hierarchical clustering method based on their spatial similarity. The clustering process relies on the Total Spatial Variance (TSV) method to measure trajectory spread, and the optimal number of clusters is chosen at the point where further TSV reduction provides minimal improvement. Each cluster represents a distinct air transport pattern shaped by synoptic-scale meteorological conditions, offering insights into the movement of atmospheric pollutants and aerosols. The classification of aerosol types is influenced by factors such as the origin and pathway of air masses, their duration in specific regions, altitude, and the location and height at which the analysis is conducted. A similar approach in this way has been made previously by Pawar et al. (2015) over Pune to obtain the aerosol types from back trajectory analysis.

Initially, we generated seven-day back trajectories during the campaign months at 500 m and 4,000 m (above and below the boundary layer, respectively) across the study locations using the HYSPLIT software. Once the multiple backward trajectories are generated, these are then grouped into clusters using the clustering tool associated with the HYSPLIT. The obtained air mass trajectory clusters are shown in **Fig. S4.** Here, each percentage defines the percentage of air mass clusters reaching a given altitude from a given direction. For example, a 50% cluster percentage means that there is a 50% chance that the air masses arriving at a given altitude over a specific location originate from that particular direction.

To associate an aerosol type with each of these clusters, the following geographic sectors were defined to identify the air mass origin at a given study location:

1. A northwest/west (NW/W) sector that includes the North African countries, Arabian Peninsula, northwestern India (Thar Desert), and other Asian countries (such as Pakistan and Afghanistan) that are expected to contribute dust aerosols.
2. A northern sector (N), mostly the IGP, with air masses containing highly polluted aerosols
3. Eastern (E) sector with the air masses mostly from the Bay of Bengal of oceanic origin
4. Southern (S) sector, with the air masses mostly of oceanic origin;
5. Central and Peninsular India (C), where the aerosols are moderately polluted in comparison with the northern sector
6. Local (L) sector in and around the study location where the types of emissions are heavily dependent on the degree of urbanization.

Once the origin of an air mass is identified for a given cluster at a specific altitude, the aerosol composition from Hess et al. (1998) that best matches it is assigned.

1. **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.;
2. **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.;
3. **Continental Average (CA)** (used to describe continental areas with moderate anthropogenic influence) assigned for the air masses originating/passing through the C sector.
4. **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
5. **Desert (DST)** to describe aerosol over the arid regions, which are assigned to the long-range transported air masses from the northwestern sector.

It is also important to note that the scale height of marine aerosols is typically small (less than 2 km); therefore, we only considered this type within the boundary layer. Additionally, air masses of oceanic origin that remain on land for more than 24 hours before reaching their destinations are classified as aerosols of continental or local origin.

The optical properties of these aerosol types for different RH conditions are obtained from the OPAC (Optical Properties of Aerosols and Clouds) model (Hess et al., 1998). **Table S3** presents the identified aerosol composition corresponding to each air mass cluster below the boundary layer (at 500 m) and above the boundary layer (at 4000 m).

After we obtained the cluster fractions of aerosol type associated with each cluster, the composite SSA and ASY at 500 m and 4000 m are computed using the following formulae:

(4)

(5)

Here, *n* is the total number of clusters, *fc* is the fraction of a cluster representing a particular aerosol type, and SSAc and ASYc denote the respective single scattering albedo and asymmetry parameter at a given wavelength and altitudinal RH corresponding to the identified aerosol type. Once these composite optical properties are computed as a function of altitude, we assigned the SSA and ASY obtained for the clusters at 500 m for the altitude bins from the surface to 2 km. For the remaining altitude bins up to 10 km, the SSA and ASY at 4000 m are used. Thus, this approach provides a realistic representation of aerosol optical properties across altitudes in the lower atmosphere (0 to 10 km) while accounting for variations in RH across the altitude bins.

**Supplementary Tables**

**Table S1**: Altitude-resolved relative humidity (RH) thresholds used for cloud screening of 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% |
| 6-10 km | 78% | 88% | 70% |

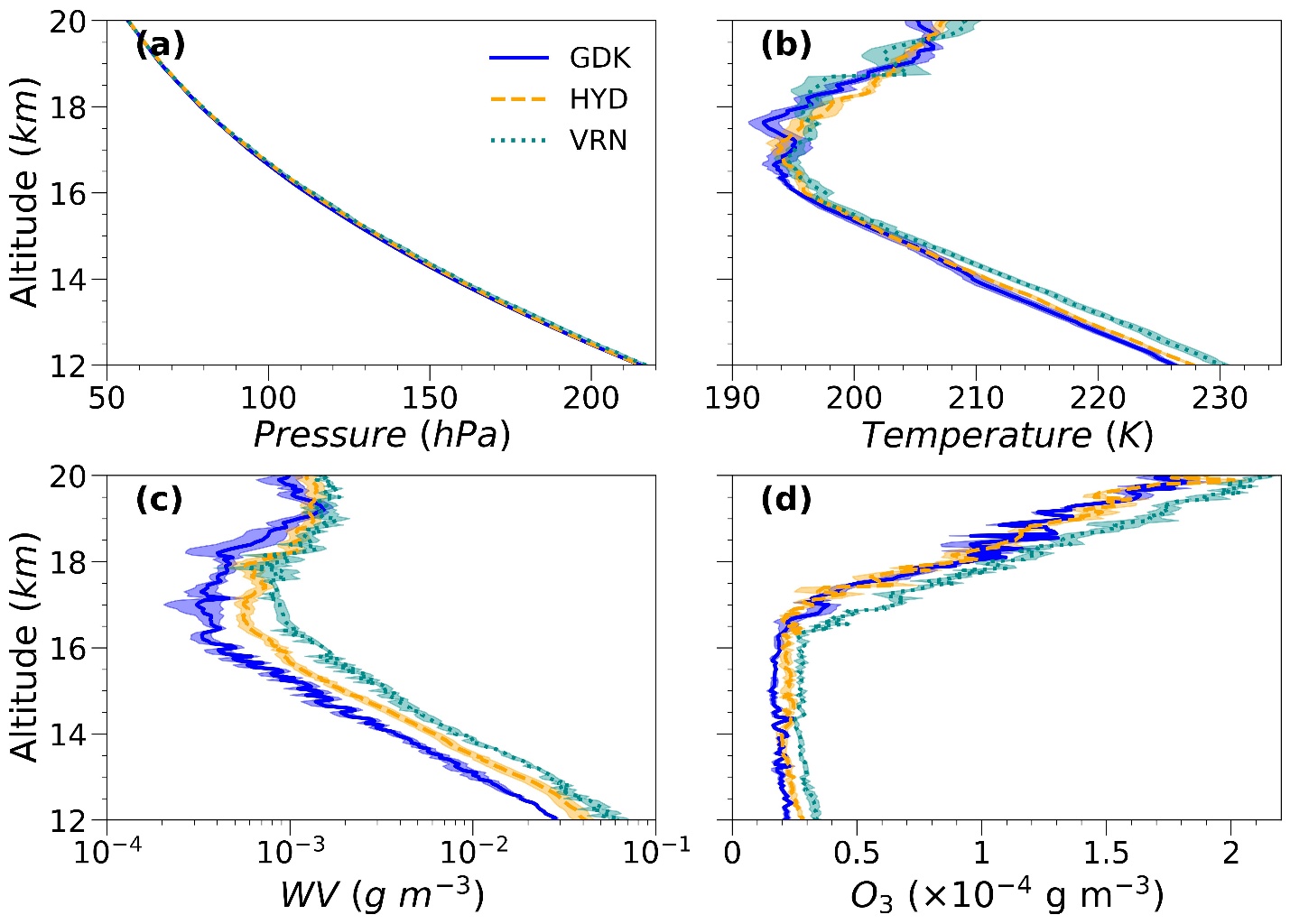
**Table S2:** Optical parameters of individual aerosol species—sulfate, nitrate, organic carbon, and ammonium—used in this study under dry conditions.

|  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Species** | **Size Range**  **(μm)** | **Mode radius**  **(μm)** | **Standard Deviation**  **(μm)** | **Refractive Index** | | **SSA** | | **ASY** | | **References** |
| **455 nm** | **940 nm** | **455 nm** | **940 nm** | **455 nm** | **940 nm** |
| **Sulfate** | 0.005-20 | 0.07 | 2.03 | 1.432-10-8i | 1.425-10-8i | 1.0 | 1.0 | 0.724 | 0.679 | Hess et al. (1998) |
| **Nitrate** | 0.5-2[a] | 0.15[b] | 1.9[b] | 1.4-10-8i[b] | 1.4-10-8i[b] | 1.0 | 1.0 | 0.685 | 0.539 | [a]Vernier et al. (2022)  [b] Zhang et al. (2012) |
| **Organic Carbon** | 0.015-0.13[c] | 0.073[d] | 1.6[d] | 1.53-0.005i[d] | 1.52-0.009i d | 0.946 | 0.65 | 0.36 | 0.084 | [c] Bossolasco et al. (2021)  [d] Jeong et al. (2020) |
| **Ammonium** | 0.015-0.62[c] | 0.05[e] | 2[e] | 1.535-10-7i[e] | 1.52-10-7i [e] | 1.0 | 1.0 | 0.457 | 0.289 | [e] Wang et al. (2022) |

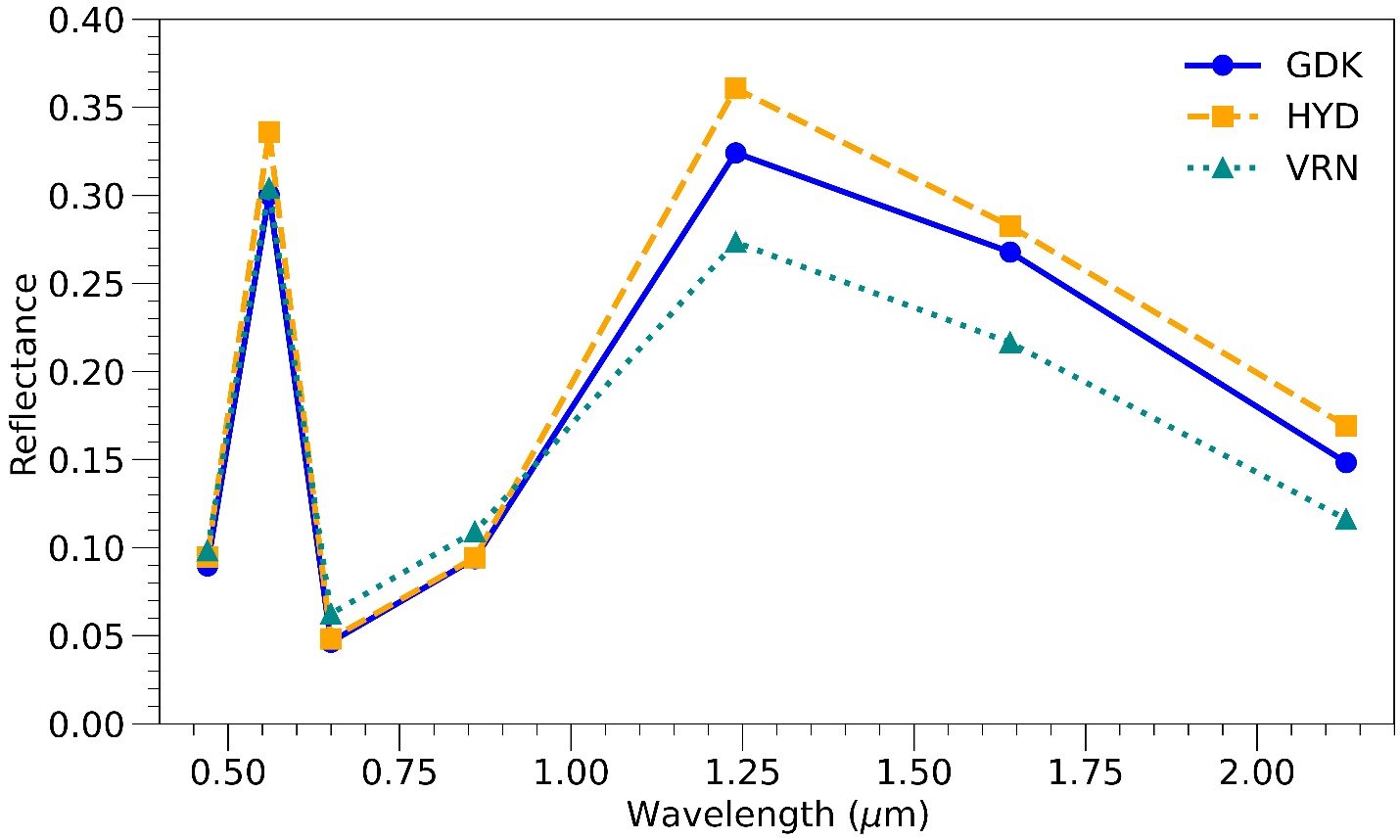
**Table S3:** Fractional aerosol composition (in parentheses) identified through cluster analysis of air mass back trajectories within and above the boundary layer at the three study locations.

|  |  |  |
| --- | --- | --- |
| **Location** | **Below Boundary Layer (500m)** | **Above Boundary Layer (4 km)** |
| Gadanki | **Cluster 1:** Maritime Tropical (62%)  **Clusters 2 and 3:** Continental Average (38%) | **Clusters 1 and 2:** Desert (73%)  **Cluster 3:** Continental Average (27%) |
| Hyderabad | **Cluster 1:** Maritime Polluted (41%)  **Cluster 2:** Urban (59%) | **Clusters 1 and 2:** Desert (56%)  **Cluster 3:** Polluted Continental (44%) |
| Varanasi | **Cluster 1:** Continental Average (20%)  **Clusters 2 and 3:** Urban (80%) | **Cluster 1:** Continental Average (51%)  **Cluster 2:** Continental Polluted (30%)  **Cluster 3:** Desert (19%) |

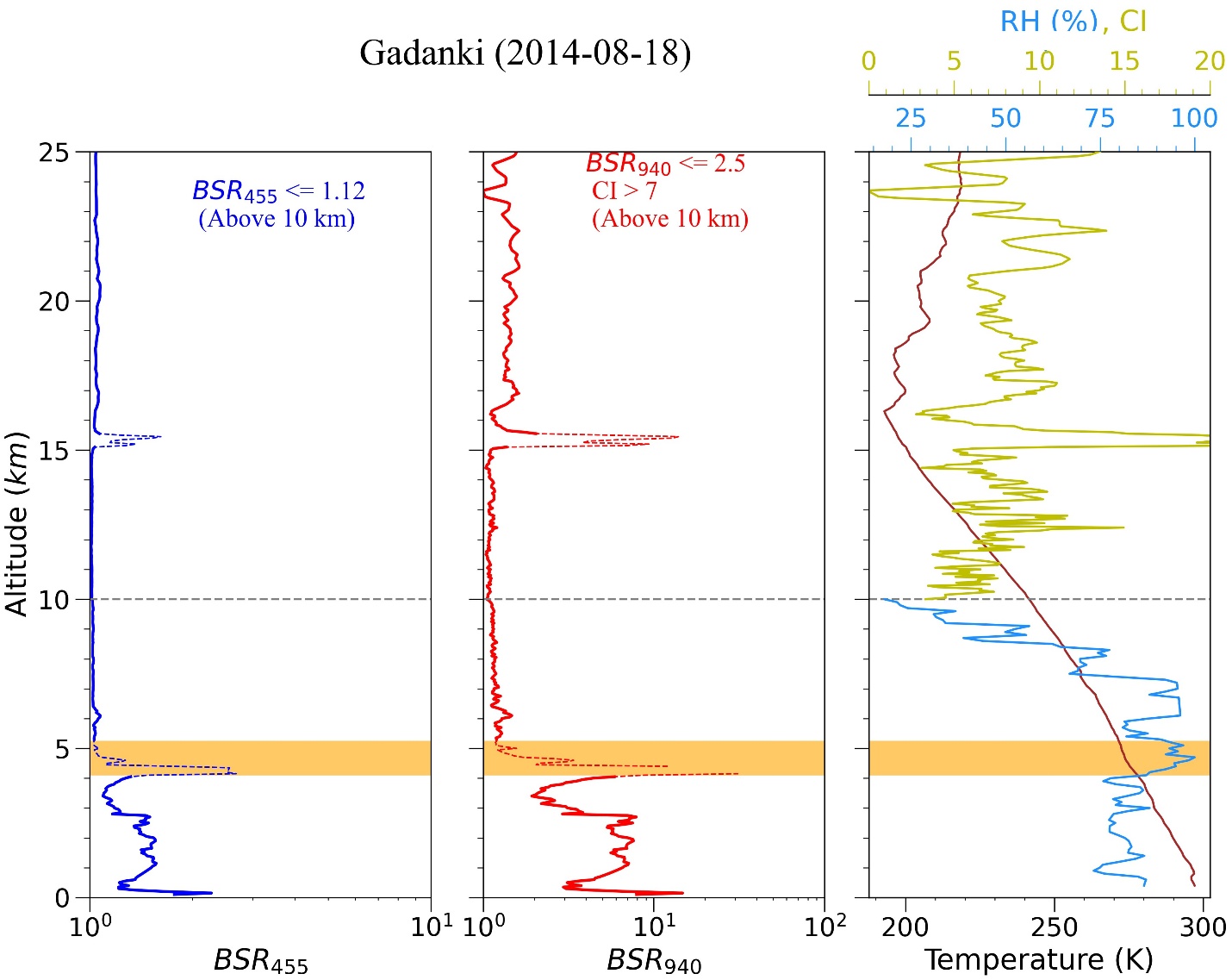
**Supplementary Figures**



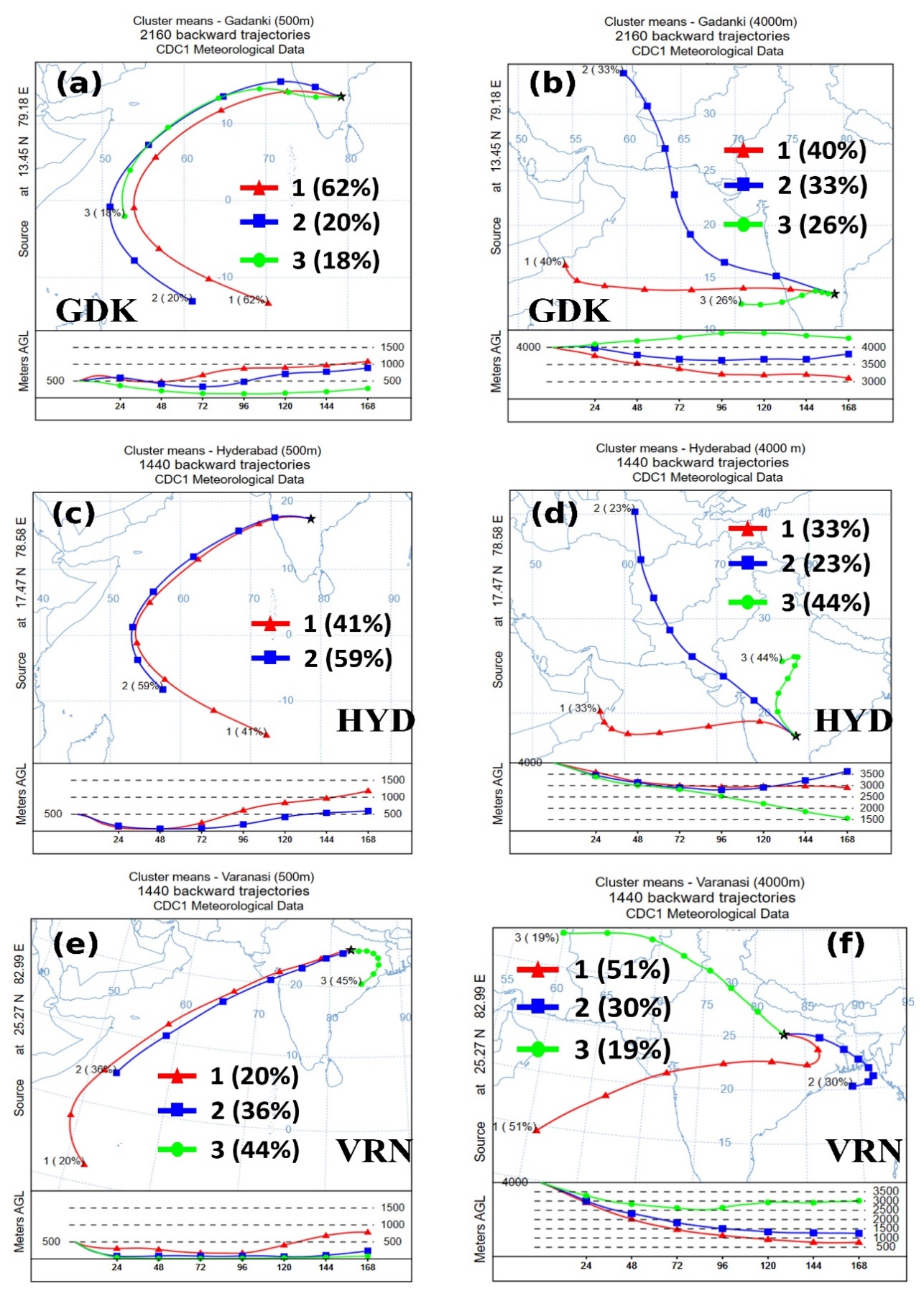
**Figure S1:** Vertical profiles of (a) Pressure, (b) Temperature, (c) Water Vapor Density (WV; log scale), and (d) Ozone Density (O₃) in the UTLS region for Gadanki (GDK), Hyderabad (HYD), and Varanasi (VRN). The log scale in panel (c) enhances the visibility of variations in water vapor concentration across locations.



**Figure S2:** Spectral variation in surface reflectance at the study locations: Gadanki (GDK), Hyderabad (HYD), and Varanasi (VRN).



**Figure S3**: Cloud screening procedure for a given COBALD profile, based on RH thresholds below 10 km and thresholds for backscatter ratio (BSR) and Color Index (CI) above 10 km. The shaded region indicates the identified cloud, while the dashed lines represent the data that has been screened out.



**Figure S4:** Seven-day air mass back trajectory clusters analyzed at 500 m and 4000 m above ground level over Gadanki (GDK) (a, b), Hyderabad (HYD) (c, d), and Varanasi (VRN) (e, f). The numbers in different colors indicate the percentage of air mass clusters arriving at each altitude from a specific direction. For example, a 50% cluster percentage signifies a 50% probability that the air mass reaching the given altitude at a location originates from that direction.

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