Regional transport of aerosols from Northern India and its impact on boundary layer height and air quality over Chennai, a coastal megacity in Southern India.

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Abstract. Anticyclonic wind circulation is prevalent over India during winter season, causing air masses to advect from the heavily polluted North India towards south-eastern coastal regions of India and the Bay of Bengal. Here, we use a synergy of satellite, radiosonde and ground-based measurements to characterize this phenomena of these regional aerosol transport event (RTE) and their impact on the boundary layer height and air quality over Chennai, a tropical South Asian megacity. The long-term satellite data and back-trajectory analysis enables us to detect occurrence of RTEs over Chennai, which can span for 2-44 days duration. The transported aerosol layer is generally located at elevated altitudes ~1-3 km along the eastern coast of India. The duration of these RTEs in winter season over Chennai accounts for ~10-15 percent5 of the days and their and their occurence is increasing since last 10 Years. Radiosonde analysis using five sites within the transport pathway illustrates distinct aerosol-associated warming (1 to 2 K) at altitudes corresponding to these elevated layers and hence strong enhancement in lower tropospheric stability during the RTEs. In agreement, the regional aerosol/ haze transport significantly reduces the boundary layer height to less than 1 km compared to background clean days (~2-2.5 km) over the east coast. Consequently, an increase in $PM_{2.5}$ concentration over Chennai is observed (~(50-55%) during RTEs compared to background days. This study provides robust observational evidence on the importance of regional transport of aerosols on air quality of downwind megacities and warrants more observational and modelling studies in future.

1. Introduction

Atmospheric aerosols are pivotal in regulating Earth's climate systems by influencing radiation budget, cloud properties and biochemical cycles. Direct and indirect effects of aerosols on the radiation balance of the Earth-Atmosphere system are evident (Comstock and Sassen, 2001; Haywood and Boucher, 2000; Lohmann and Feichter, 2005; Satheesh and Krishnamoorthy, 2005; Yu et al., 2006) and it is believed to generate climate perturbations on a regional and global scale. Apart from the local generation, the long-range transport of aerosols from their sources can severely pollute a large area far from the apportionment and it is mainly influenced by the atmospheric circulation and aerosol lifetime. Although local emissions contribute mainly to hazy episodes in megacities, it can also be influenced by regional pollutant transports (Ma et al., 2020; Mhawish et al., 2022). Such hazy events can cause severe air pollution, adversely affecting public health. Prolonged haze events and associated high $PM_{2.5}$ loading have frequently been reported over South Asia and China during recent autumn and winter

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seasons (Qin et al., 2016; Yang et al., 2020; Zhang et al., 2021a). The significant factors influencing such hazy events were attributed to stable synoptic conditions with weak surface winds and low Atmospheric Boundary Layer Height (ABL-H) (Wang et al., 2014) along with the regional aerosol transport and Atmospheric Boundary Layer (ABL) interaction (Zhang et al., 2015).

Such transported aerosol layers, stratified above the ABL, can significantly affect the surface energy balance and ABL dynamics owing to their interaction with incoming solar radiation (Ding et al., 2016; Ma et al., 2020). Depending on the dominant aerosol species, the net impact of these layers could be absorbing or scattering of incoming solar radiation. In either case, the presence of this transported aerosol layer can induce cooling at altitudes below the layer and warming around and above the altitudes where they are located. Simultaneously, near-surface accumulation of absorption aerosol concentration (under a shallower boundary layer) can lead to lower atmosphere warming and surface cooling. Thus, a series of thermodynamical effects can ensue disrupting stability and enhancing the upward transport of heat and aerosol through turbulent motion (Barbaro et al., 2014; Huang et al., 2018). In continuation, previous studies found the role of aerosol on the suppression of ABL development through their relative heating and cooling in the upper atmosphere and surface, respectively (Liu et al., 2019; Petäjä et al., 2016; Wang et al., 2019b, 2020, 2018; Wilcox et al., 2016; Zhao et al., 2019; Zou et al., 2017).

Hence, understanding and characterising the regional transport of aerosols on the ABL structure and air quality are complex. There are studies signifying the role of aerosols on the boundary layer dynamics (Aruna et al., 2013; Huang et al., 2018; Ma et al., 2022; Miao and Liu, 2019; Raatikainen et al., 2014); however, most of them are based on the modelling framework, and observational evidence is scarce. This study aims to delineate, for the first time, the effects of transported aerosols from north India towards the southern part of the Indian peninsula on the boundary layer dynamics and hence the pollution dispersion using collocated high-resolution lidar, radiosondes, surface weather observations along with space-based observatories.

The Indo-Gangetic Plains (IGP), the densely populated and growing economy of the Indian subcontinent, experiences high aerosol loading both around the surface and in the vertical column during the winter season attributed to the wide range of anthropogenic activities ranging from biomass, fossil fuel burning and agricultural activities (Prasad et al., 2006; Ramanathan and Ramana, 2005; Tripathi et al., 2006). The prevalence of a high-pressure system over the central Indian landmass, especially during the winter seasons (December to March), generates a persistent northeasterly offshore flow (Krishnamurti et al., 1998). It provides a pathway for transporting aerosols from continental areas into the otherwise pristine ocean, covering thousands of kilometres in less than ten days (Krishnamurti et al., 1998; Rajeev et al., 2000). As such, pollutants from North India can get transported to the Bay of Bengal and then towards South India under the influence of prevalent strong convection and anticyclonic cyclonic circulation formation over the northwest of the Bay of Bengal (Prijith et al., 2016; Rajeevan and Srinivasan, 2000). Such transboundary transport of pollutants is evident in widespread pollution over the southern Indian peninsula (Ananthavel et al., 2021b; Kant et al., 2023; Mehta et al., 2023; Mhawish et al., 2022; Ratnam et al., 2018; Thomas et al., 2021). There is a campaign-based investigation held over the Indian Ocean, e.g., the Indian Ocean Experiment (INDOEX) (Ramanathan et al., 1995) to investigate the characteristics of transported aerosols towards the Indian Ocean and the Arabian Sea (Chester et al., 1991; Prodi et al., 1983; Savoie et al., 1989). The studies revealed that the transported aerosol predominantly consists of black carbon,

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organics, sulfate, nitrate, ammonia, sea salt, and mineral dust (Ramanathan et al., 2001). An increase in the aerosol loading in the free troposphere reduces the amount of incoming solar radiation reaching the surface, thus causing dimming while warming the mid and upper troposphere and cooling the surface (Dipu et al., 2013; Sarangi et al., 2018). On the other hand, they significantly alter the atmosphere's underlying thermodynamics, leading to modifying the boundary layer structure. Hence, it is essential to characterise such transports, especially their occurrence characteristics and the nature of the aerosols present. However, observational evidence on such transboundary aerosol transports, their frequency of occurrences, their impact on the ABL development and the regional pollution maintenance have not been attempted yet; this study primarily focuses on unravelling such aspects.

Here, long-term satellite observations from Moderate Resolution Spectroradiometer (MODIS), Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observatory (CALIPSO) and back-trajectory analysis are used to understand better and characterise the spatiotemporal variability in long-range regional transport of aerosols from North India to central-southern India during the winter season. Further, we have also used ground based observations of Micro Pulse Lidar, Radiosonde, surface weather and surface $PM_{2.5}$ measurements to (i) investigate and characterize these regional aerosol transport episodes over east coast of India , (ii) quantify the associated changes in the air temperature profiles, lower tropospheric stability, ABL-H over the east coast region and (iii) quantify the associated changes in the surface $PM_{2.5}$ distributions due to ABL-H reduction over Chennai. Section 2 describes the datasets used, followed by the methodology for composite analysis of aerosols during the Regional Transport Episodes (RTE) days and clear days. Further, results and discussion are provided in section 4 and the conclusion in section 5.

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2. Dataset and Methodology

Space-based observations

MODIS on board the polar orbiting sun-synchronous satellites (Terra and Aqua) is utilised to estimate the aerosol optical depth (AOD) information at 550 nm. The MODIS measures radiance at 36 spectral bands in the visible to thermal IR spectral range of 0.41-14 µm (Kaufman et al., 1997). Within the spectral range, 7 bands are dedicated for aerosol measurement having a spatial resolution of 250m/500m. Owing to its large spatial swath (2330 km), MODIS is capable of observing the entire globe in a single day during two different times, i.e., at 01:30 AM/PM (Aqua) and 10:30 AM/PM (Terra) local time, which crosses the equator. We used the current version of Multiangle Implementation of Atmospheric Correction (MAIAC), which retrieves the AOD over land and ocean at 1 km resolution (Lyapustin et al., 2011b, 2011a), between December and March during 2015-2024 in this work.

In addition, the space-based lidar observation, Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP, (Winker et al., 2009; Young et al., 2013) onboard CALIPSO is utilised to understand the vertical variation of aerosol extinction profiles. The level 2, 5 km (horizontal averaged) standard aerosol profile (AProf) version 4.51 at 532 nm during December – March between 2015 and 20233, segregated during the RTE and clear days, are used. The CALIPSO crosses the equator ~01:30 AM/PM; we used both the day and night passes, around ± 5 degree over the eastern coastal box (as shown in Fig.1a), for the present study.

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In situ observations

The Micro Pulse Lidar (MPL), an elastic backscatter dual-polarization lidar of Droplet Measurement Techniques (DMT, USA), is located at the premise of SRM IST (12.80N,80.0E, 45m above mean sea level). The instrument is set up at the Atmospheric Observation and Modelling Laboratory (AOML, 40m above the ground level), at a total height of 85m above mean sea level. The Normalised Relative Backscatter (NRB), between January and February during 2018 and 2023, is primarily utilised to retrieve total attenuated aerosol extinction and determine ABL-H. Details on site description and technical specifications about the MPL (Ali et al., 2022), retrieval of extinction coefficient and AOD (Ananthavel et al., 2021a, 2021b) and ABL-H estimation (Kakkanattu et al., 2023; Reddy et al., 2021a) are provided in references.

The diurnal variability of ABL-H from MPL is estimated using the Wavelet Covariance Transformation (WCT) method (Baars et al., 2008; Davis et al., 2000; Pal et al., 2010; Reddy et al., 2021a), which estimates the ABL-H from lidar profiles by step changes in signals using Haar function. We have also identified the top of the transported aerosol layer (The TAL) using the differential zero crossing method (Ali et al., 2022; Mehta et al., 2023), similar to the methodology followed by (Mehta et al., 2023) to identify the elevated aerosol layer. In general, the extinction coefficient gradually decreases above the ABL. However, presence of TAL can increase the extinction values similar to as observed within the ABL. The differential zero crossing method identifies the top of the TAL using the gradient of extinction coefficient profiles. Note that, this method of TAL detection is used only when a valid ABL is identified.

Upper air and surface weather information used in this study are obtained from India Meteorological Department (IMD) from the sounding over Kolkata (22.65N, 88.45E, 6m above MSL), Bhubaneshwar (20.25N, 85.83E, 45.0m AMSL), Vizag (17.68N, 83.33E, 69.9m AMSL), Chennai (Meenambakkam) (13.0N,80.06E, 16m above MSL), about 20.13 km northeast of SRM IST, Kattankulathur and Karaikal (10.9N,79.8E, 6.9m above MSL). The radiosonde data archived at 05:30 LT between December-January 2015 and 2024 are used to interpret the meteorological conditions during the aerosol transport periods and ABL-H determination. The ABL-H is estimated from the potential temperature profiles, obtained from the Radiosonde. The ABL-H is the altitude at which the maximum potential temperature gradient observes at the lower troposphere.

Hourly $PM_{2.5}$ measurements are routinely made at the U.S. Embassy and Consulate, Chennai using a beta attenuation monitor (San Martini et al., 2015). The dataset within the study period (December – January 2015 and 2024) is obtained from AirNow (http://www.airnow.gov). The $PM_{2.5}$ observations from the U.S. Embassy are validated and in good agreement with other observations (Jiang et al., 2015; Mukherjee and Toohey, 2016). The datasets are used to investigate the distribution of surface pollution during the haze transport from IGP to Chennai.

Reanalysis datasets and back-trajectory analysis

The Modern Era Retrospective analysis for Research and Application, Version 2 (MERRA2) is employed to understand the spatial variation of Total Aerosol Extinction (TAE), radiation flux and, wind parameters (U, V and resultant wind speed ($\sqrt{U^2+V^2}$)). The MERRA2 reanalysis product provided by NASA's Global Modeling and Assimilation Office (GMAO) is available at 0.5° x 0.625° spatial resolution (Gelaro et al., 2017). MERRA2 simulates five aerosol species, including sulfate, black carbon, dust, organic carbon and sea salt, with Goddard Chemistry, Aerosol, Radiation and Transport (GOCART) model and their simulated properties are found to be robust (Randles et al., 2017). We used datasets between December and March during 2015-2024.

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The Aerosol Direct Radiative Forcing (ADRF) at the surface is estimated from the radiative fluxes, provided in the MERRA2 product 'M2T1NXRAD', by taking the fluxes with aerosols and without aerosol under clear sky condition. The variables SWGNTCLR (surface net downward shortwave flux assuming clear-sky), SWGNTCLRCLN (surface net downward shortwave flux assuming clear-sky and no aerosol), LWGNTCLR (surface net downward longwave flux assuming clear sky) and LWGNTCLRCLN (surface net downward longwave flux assuming clear-sky and no aerosol) are used to calculate ADRF at the surface (Thomas et al., 2019, 2021).

ADRF = (SWGNTCLR + LWGNTCLR) – (LWGNTCLR + LWGNTCLRCLN)

The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) Model is used to compute the air mass backward trajectories (Rolph et al., 2017; Stein et al., 2015). We used web-based HYSPLIT model (https://www.ready.noaa.gov/HYSPLIT.php) to estimate the 5- day backward trajectories reaching Chennai, to understand the pathways of aerosol transport from northern India. The Global Data Assimilation System (GDAS) meteorological data sets at a spatial grid of 1.0° were used as input to simulate the air mass backward trajectory. The trajectory height was set at every 0.5 km between surface (50m) and 4.0 km, with backward trajectories calculated at 6–hr intervals for the aerosol transport days and clear days. The trajectory density was determined by counting the number of trajectories crossing each grid cell separately.

Methodology

Composite analysis of RTE and Clear days

The RTE days are, by definition, the days with significant aerosol transport from North India to south-coastal India, inducing a widespread haziness over the eastern coastal box. Hence, the daily AOD values over the East Coast and nearby Bay of Bengal (black box in Fig.1a), referred to as "eastern coastal box" is expected to have extreme values during RTE days. The variability in MODIS-observed daily AOD values within the east coast box was analysed and it was found that AOD values equal to 0.7 and 0.3 represent the 70th and 30th percentile values, respectively. Accordingly, RTE days are the extreme event days when domain mean AOD is greater than 0.7. Conversely, the days when mean AOD values averaged within the east coast region are less than 0.3 are classified as 'clear' days. Manual inspection of all the identified days (RTE and background) was done for confirmatory check of the visible long-range aerosol transport from the IGP towards the south Indian peninsula is observed (similar to the case shown in Supplementary Fig. S1 for a reference). Many days in both composites has cloud contamination present, hence, the cloudy days over east coast box (i.e days with mean cloud fraction > 0.1) are excluded from the composites of both RTE and Background clean days. Based on both these criteria, we have 119 number of RTE days and 71 number of background clean days for analysis. We used these segregated days to perform composite analysis toto understand the RTE associated perturbations in aerosol vertical distribution (CALIPSO and MPLMPL observations), temperature profiles and ABL-H (IMD Radiosondes) and MERRA-2 simulated Total Aerosol Extinction (TAE), surface net radiation fluxes and aerosol radiative forcing over the east coast of India (compared to background clean days). CALIPSO swaths available around the eastern coastal box (as shown in Fig.1a) are grouped with respect to the RTE and clear days to study the three-dimensional variation of extinction profiles pertinent to the RTE phenomena. Moreover, MPL (vertical profiles of extinction coefficient)

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and $PM_{2.5}$ measurements available during RTE and clear days over Chennai are also segregated into these composite days to infer the ABL-H variability and pollution concentration. Details of the sample available for the analysis are presented in Supplementary Table 1.

3. Results and Discussion

3.1 Occurrence of RTE and clear days

The mean spatial distribution of columnar AOD over the Indian subcontinent for RTE and clear days composites are compared in Fig. 1a and 1b. The significance of regional aerosol transport from north India to south is evident, as a majority of grids in the east-coastal region and a considerable portion of the Bay of Bengal has very heavy aerosol loading (AOD > 1). In contrast, the composite mean of AOD during clear days (Fig. 1b) is substantially lower (0.23±0.06) over the eastern coastal box, as portrayed in Fig.1b; The spatial distribution of aerosols during the RTE and clear days is further confirmed using the MERRA2 reanalysis products. The average of the MERRA2 simulated columnar AOD at 550 nm superimposed with the wind vectors at 850 hPa, for the same composite of RTE and clear days are compared (Figure 1c-d). The analysis shows a similar pattern as observed from AOD distribution from MODIS data; however, variation in the magnitudes is present. Notably, the MERRA2 simulated columnar AOD values span from 0.6-0.9 on RTE days across the East Coast and is largely 50-60% greater compared to clear days.

The composites also show that the aerosol transport from the Indo-Gangetic Plain to the southern Peninsula via Bay of Bengal is predominantly by the divergence associated with the anticyclonic circulations prevalent during the winter season. The MERRA2 simulated wind circulation flow manifests into a northerly wind as it enters the Bay of Bengal and eventually merges into the easterly circulation (prevalent around the tropics) as the wind enters back into the southern Peninsula, south of 20°N. During the winter and pre-monsoon season, the westerlies wind system is prominent over Northern India and IGP in the lower troposphere prevalent to the high-pressure system generation over central India (Krishnamurti et al., 1998). It is worth noting that the easterly wind speed across the southern BoB is stronger during clear days than on RTE days. As such, the difference between the RTE and clear-day wind speed composites is that the wind speed during RTE days across the entire eastern half of the Indian subcontinent is weaker than the clear days (Supplementary figure Fig S3). The reduced wind speed is expected to promote the endurance of aerosols over the eastern coast column and induce greater AOD over the south-eastern coast and the southern Indian peninsula.

We further checked the wind back trajectory model to better understand the pathway characteristics. Fig.1e and f shows the number density of HYSPLIT trajectory analysis for the RTE and clear day, respectively, between surface (50m) and 4 km, illustrating the 5-day backward trajectories reaching Chennai, computed for every 6-hour interval. It also indicates that pollutants are predominantly transported from the northern parts of India and from the IGP outflow region over Northern BoB during RTE. In addition, there also observed transport from inland areas of the eastern coasts along the anti-cyclonic circulation pathway of the aerosol flow from North India. On the other hand, the transport is predominantly from the nearby oceanic region during the clear days. Thus, this 5-day back-trajectories also confirm the fidelity of the RTE composites and that they are indeed characteristically different from the composite of background days.

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We also checked for diurnal pattern in the MERRA2 simulated TAE values. However, such variations are negligible, pointing to the longer duration of such events. Hence, we further examined the endurance of RTE days. Normally, the RTE events prolong for days; persistence of such events for more than a day is observed to be ~53% of total observation, while 21% of occurrences have RTE durations of more than 4 days. Duration of RTE episodes, in general, can vary from one day to 4-6 days. Overall, we have categorized RTEs as episodes of varying duration, i.e. 1 day, 2 day and so on till 5 days, and the occurrence of RTEs episodes within each category at yearly basis is recorded. Supplementary figure Fig.S2S2 provides an overview of occurrence of RTE events under each category between 2015 and 2024. On average, the RTE which occurred over the south-eastern coast box for consecutive 2-4 days is the highest (as shown in Fig.S2), and such events show an increasing pattern. Notably, the year 2022 experienced a 12-day consecutive haziness between the 20th and 31st of March. Figure S2 also suggests an increasing trend in the overall occurrence of RTE days. Endurance of such hazy periods can result in significant consequences on the air quality and boundary layer dynamics.

3.2 Vertical aerosol structure during RTE and clear days

The CALIOP observed mean vertical distribution of the aerosol extinction over the south-eastern coastal region during the RTE days and clear days is shown in Fig.2a and b, respectively. As expected, there is a distinct decreasing gradient in aerosol extinction values between surface and 2 km altitude as we move from IGP in the north to southern peninsular coastal India during both RTE and clear days. More interestingly, high values of aerosol extinction (> 0.2) are discernible up to 5 km during RTE days over the region south of 20°N during RTE days, while the same is confined to altitudes less than 1.5 km during clear days. Also, over region north of 20°N, a relative increase in the extinction within ~1-4 km can be observed on RTE days relative to clear days. Note that, the averaged profiles cover both the land and ocean parts and there can be differences in the extinction coefficient due to the contrast between the land and ocean swaths of the CALIPSO. To understand the variations in the vertical distribution of aerosols between the RTE days and background days composites, we analysed the CALIPSO profiles over the land and sea pixels of the east coast box, separately (Figure S7, S8 and S9). The mean extinction during the RTE days is observed to be greater than 0.2 over the land, however it observes between 0.1-0.2 over the ocean region. While the land regions have high aerosol concentration near the surface, the same is not seen over ocean as there is no active surface emission sources over Ocean. However, the transported elevated aerosol layers are present at altitudes 1 to 3 km over both the land and the ocean regions.

Further, ground based MPL-observations over Chennai is used to study the RTE-associated diurnal and daily-scale perturbations in vertical distribution of aerosols over this region. We show the temporal changes in the vertical characteristics of aerosol extinction and TAL during a one week RTE episode between 23rd-29 January 2018 over Chennai (as shown in supplementary Figure Fig.S1 and S1A), as observed by MPL, is provided in Fig.2c. The temporal variation of background surface meteorology, including surface T, wind speed (WS), PM2.5 and the AOD, are provided in the Supplementary Figure Fig.S4. A significant increase in the columnar AOD between ~0.4 and 0.8 is observed during the hazy events. However, it maintains ~0.2-0.3 during the clear days. It is also worth noting that the AOD above the ABL (integrated extinction within the free troposphere) is observed to be close to the AOD values during the RTE period, suggesting a dominant presence of TAL above the ABL. The occurrence of the TAL can be seen above the ABL during the RTE periods and persisted for ~ 3-4 days. The top of the TAL was observed initially at ~2.5 km at ~06:00 LT (or IST) on 24 January 2018, which gradually

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reduced to ~1.5 km and merged with the ABL at 09:00 LT on 28 January 2018. Thus, the vertical distribution of total attenuated aerosol extinction between 24th-27th January 2018 is representative of RTE and the background days (23 and 28-29 January) as clear day conditions.

The study further extends to the variations in the aerosol extinction during RTE and clear days. Figure 2d shows the day averaged profiles of RTE (24 -27 Jan 2018) and clear days (23, 28, 29 Jan 2018) observed during the typical case from MPL observations, as shown in Fig.2c. Although the extinction values are observed to similar near the surface, it rapidly decreases till ~ 0.8 km during the RTE days. Further, it maximizes within the altitude range ~1-2.5 km. Overall, the aerosol extinction during the RTE days is observed to be 50-60% higher than clear days between 1-2.5 km, suggesting the presence of TAL. Fig.2c also superimposed with the ABLH determined from MPL observations and temperature profiles obtained from radiosonde observation from Chennai (Meenambakkam). The observed temperature profiles indicates the top of the TAL. Interestingly, the ABL-H also decreased from ~1.4 km to ~0.3 km (~78% reduction) between 24 and 25 January 2018. The mean ABL-H is observed to be 1.3±0.8 km and 1.8±0.8 km during the RTE and clear days, respectively, exhibiting an overall reduction of ~40%. Temperature inversions are also observed near the top of the TAL during the RTE, which can also be attributed to the aerosol-induced warming of the atmosphere and large-scale circulation (Ganguly and Jayaraman, 2006; Sinha et al., 2013). However, such as aspects are not addressed here due to the limited datasets.

3.3 RTE-associated atmospheric warming, lower tropospheric stability and ABL-H suppression

To understand the effect of the transported aerosols and their vertical extent on the background thermal conditions, we also analysed the vertical temperature (T) profiles obtained from radiosonde observations: Kolkata, Bhubaneshwar, Vizag, Chennai and Karaikal located in the east coast of the Indian peninsula, as depicted in Fig.3a. The average T profiles with standard error obtained for the RTE (red) and clear (blue) categories are shown separately for different stations in Fig.3b-f. The relative difference (in percent) between the temperature profiles of RTE and clear days, from the RTE ($\frac{T_{RTE}-T_{Clear}}{T_{RTE}} \times 100\%$)% is shown on the top axis as dashed lines.

Over Kolkata, where it lies in the northmost region, the relative enhancement in the temperature RTE ($\frac{T_{RTE}-T_{Clear}}{T_{RTE}} \times 100\%$) forms a parabolic shaped structure which was located between 0.3 km to 2 km and has its centre around 1 km and sharply decreases afterwards. Over Vizag and Bhubaneshwar, the intermittent stations, similar parabolic shape in relative differences is observed, spread between 0.2.5-2.5 km with centre between 1 km to1.5 km, km. Interestingly, Chennai and Karaikal, the south most stations, also observedobserves the parabolic shape heating starting 0.5 km but the upper stretch was extended up to 3 km, with centre located between 1.5 km to 2 km. This phenomenon also suggests that aerosol-induced warming at the lower troposphere not only enhance the temperature at the altitude where aerosol occur but also modifies the overall temperature profiles of the lower atmosphere. Moreover, the observed phenomena of enhancement in altitude of RTE-associated warming as we move southward from IGP, is also consistent with the fact that the long-range transported aerosol plumes gets elevated at downwind locations (Stohl, 2006; Yu et al., 2012)(Stohl, 2006; Yu et al., 2012). Over Chennai, the altitude of observed peak warming coincides with the peak occurrence of TAL, as observed in Fig.2c, suggesting the aerosol-induced radiative effects of TAL on temperature profiles. Also, the observed warming occurs throughout the column till ~ 3km over both Chennai and Karaikal. This phenomenon also suggests that aerosol-

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induced warming at the lower troposphere not only enhance the temperature at the altitude where aerosol occur but also modifies the air temperature near surface. Similar to the observation over Chennai, the observed heating during the RTE days over the other stations can also attributed to the presence of TAL. However, the observed latitudinal differences in the magnitudes of warming can be due to the spatial inhomogeneity of the aerosol concentrations, mainly attributed to the transport strength. Interestingly, the intensity of RTE-associated warming over megacities like Kolkata, Bhubaneshwar and Chennai are greater than that of Vizag and Karaikal, suggesting role of local emissions.

Figure 4a examines the relative differences in lower tropospheric stability (LTS i.e. gradient in the temperature between 1.5 km and surface (ΔT)) during the RTE and clear cases across the different stations over the eastern coast. During the background clear days, the LTS is largely ~ ~7K but the same during RTE days is skewed ~ 4K suggesting the enhanced LTS and hence atmospheric stratification. The variations in the LTS can have stronger impact on the ABL-H, and the relative suppression of ABL under the influence of the TAL are addressed in Fig.4b-c.

For ease of comparison and increase the sample size, we grouped radiosonde-estimated ABL-H data over Kolkata, Bhubaneshwar and Vizag (representing the north region of the east coast box) into one comparison plot (Figure 4b), and the observed ABLH over Chennai and Karaikal into the other plot (representative of transported plume to South region of east coast box). It is also interesting to observe a latitudinal heterogeneity in the peak occurrence of ABL-H over the east coast. The mode of distribution of ABL-H during clear day composite varies between 1.5 – 2 km over north region and the same is 2 -2.5 km over the south region. During RTE days the distribution of ABL-H shifts drastically to lower values and the mode of distribution is decreases significantly to ~0.3-0.4 km over north region and 0.5 -1 km over south sites suggesting the strong influence of TAL-associated LTS on suppression of ABL across the eastern coast. As an effect of the accumulated aerosol concentration above the ABL heats the thermal inversion layer and strongly suppresses the ABL development (dome effect) (Ma et al., 2020). The shallow ABL further promote severe hazy episodes (Quan et al., 2014; Ye et al., 2016).

Figure 4d. shows the mean ABL-H observed over SRM IST (Chennai) during the RTE (red, 10 days) clear days (blue, 6 days). Overall, the mean ABL-H observed during clear days and RTE days are observed to be ~0.9±0.3 km and ~0.6±0.2 km, respectively, accounting a relative suppression of ABL-H by ~33-35% during RTE days. Notably, such decrement in the ABL-H was dominant during the afternoon hours (15-19 LT), where it shows a declined from clear days (1.0±0.1 km) to RTE days (0.6±0.1 km) by ~67-70%. Such dominant suppression was also observable during forenoon hours (08- 12 LT), accounting a reduction of ~45% (2c and e). Moreover, the ABL-H evolution is also considerably different between the two composites. While the evolution of ABL-H with time of the day is gradual during the clear days, the transition of ABL-H between midday and afternoon is much steeper during RTE days. Also, the time to reach maximum ABL-H is delayed by ~1-2 hours during RTE compared to background clear days. The suppression of the ABL followed by accumulation of absorptive aerosol in the upper ABL has been investigated earlier, especially through numerical simulations (Ding et al., 2013, 2016; Zhao et al., 2019); however, observational evidence is scarce. (Barbaro et al., 2014) suggests that a drop in the ABL from 1.4 km to 0.9 km (~35% reduction), through sensitivity experiments. Similarly, (Wang et al., 2015) suggest that the stable stratification of the atmosphere above the ABL through the significant warming by absorbing aerosols contributed to a decrease of ABL-H by 33%. Observational studies over China suggest that the occurrence

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of elevated aerosol layers has induced suppression of ABL-H from 1.27 km to 0.78 km (~38% reduction) hence lifting the surface pollution level to 118% (Wang et al., 2018). (Zhang et al., 2021b) reports a reduction in the ABL-H from 1.09 km to 0.48 (~60% reduction) during intense haze episodes over China.

Supplementary figure Fig. S6 shows the aerosol direct radiative forcing (ADRF) observed during the RTE and clear days, estimated from MERRA2 radiative flux observations at the surface (Thomas et al., 2019, 2021). Overall, the ADRF has a net cooling at the surface both during the RTE and clear days. However, RTE triggers to enhance the cooling of the surface to ~-20-40 W/m^2. Such strong cooling observes to be around the eastern coastal regions where the aerosol transports generally occur. In specific, it reduced to less than -40 W/m^2 over the eastern coastal box where the TAL present. During clear days (Fig.S6b), the strong cooling is confined over the IGP alone (~-25-30 W/m^2). The difference in the ADRF during the RTE and clear day composite is shown in Fig.S6c, evidencing a cooling of ~-15-20 W/m^2 by the RTE days. It also suggests that aerosol transport from the north India has a profound effect on the radiation and eventually on the ABL-H. The solar dimming due to the presence of TAL can block the solar radiation reaching the surface, resulting in the overall dimming in the ground surface, weakening the surface flux, perturbing the convective process and suppressing the ABL development. The development of ABL during daytime is mainly dominated by convective processes (Garratt, 1994; Stull, 1988); however, the formation of TAL suppresses such development, via reduction in the incoming solar radiation, especially the surface dimming and inducement of the "dome effect"." (Guo et al., 2017; Petäjä et al., 2016). Enhancement in the suppression of ABL-H distribution during the afternoon hours can also be attributed to the thermal internal boundary layer formation, where the transport of pollutants towards the land from BoB under the influence of sea breeze (Reddy et al., 2021b). However, this contention requires further investigation with large samples.

3.4 Impact of transported aerosols on the air quality of downwind megacities

The association of ABL-H and $PM_{2.5}$ is delineated using collocated observations of MPL and $PM_{2.5}$ measurements over Chennai using the measurement across SRMIST and US Consulate, Chennai, respectively. Fig.5a5a shows the scatter of normalised anomalies between $PM_{2.5}$ and ABL-H during RTE days, showing for each 2 percentile observations. Note that, the first and last 4 percentile of the observations are excluded from the analysis to remove outliers. The normalised anomalies are obtained by subtracting the parameters from the climatological average (during the winter season) and further divided with the same ($X\ Anom. = \frac{X-X_{Mean}}{X_{Mean}} \times\times 100\%$, where X = $PM_{2.5}$, ABL-H). Note that, average ABL-H during the winter season is estimated with the NRB during 2018 and 2023 alone. However, it matches with the climatological ABL-H estimated by (Reddy et al., 2021a) over Chennai. As expected, the normalised anomalies between $PM_{2.5}$ and ABL-H are negatively related, with a statistically significant (>95% confidence) correlation of -0.6. It portrays that, overall, a 30% reduction in the ABL-H can contribute to ~130-150% increase in the surface $PM_{2.5}$ concentrations. Observational studies by (Su et al., 2020) show a nearly similar relationship during COVID-19 in China; however, with a different correlation value over Beijing and Northern China, attributed to the inhomogeneity in the spatial distribution of pollution.

Finally, the overall diurnal changes in $PM_{2.5}$ during the RTE and clear day composites are portrayed in Fig.5b5b. As expected, the RTE days experience ~50-55% enhancement in $PM_{2.5}$ than clear days. During the clear

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days, the $PM_{2.5}$ increases steeply during the early morning hours and maximizes at 50 µg/m^3 at 08:00 LT. This is followed by a gradual decrease to the daily minimum value of ~ 20 - 25 µg/m^3 from 08:00 till 15:00 LT, before increasing slightly in evening and night till 30 µg/m^3. In comparison, the RTE composite mean shows similar diurnal variability in the $PM_{2.5}$ values but the magnitudes are greater. The maximum value at 08:00 LT is ~ 80 µg/m^3 and the daily minimum value is ~ 35 µg/m^3 at 15:00 LT, followed by slight increase in evening ~ 40µg/m^3. As such, the percentage increase in $PM_{2.5}$ during RTE composite is more than 50% with the maximum being around morning 0900-1100 LT of ~65-80%. This is probably due to shallower ABLH over the megacity of Chennai during winter. Note that Fig.4c illustrated the decrease in ABLH is maximum during morning and evening time and least during midday. The clear days, characterized by elevated ABL-H, often promote enhanced wind speed and vertical mass movement (Xiang et al., 2019) can result reduction in the surface pollutions. On the other hand, a suppressed ABL-H as observed during the RTE days significantly affect the vertical dispersion, leading to higher concentrations of pollutants near the surface (Wang et al., 2019a). It is also to be noted that, the surface pollution aggravation becomes complicated in the presence of stable boundary layer, such that, it hinders the exchange of pollutants and energy between the surface and free troposphere and potentially leading to higher concentration of pollutants in the atmosphere if they are not cleared otherwise (Shi et al., 2020).

4 Summary and Conclusion

This paper presents the first observational evidence of the effect of long-range transported aerosols on the boundary layer dynamics and $PM_{2.5}$ enhancement over the east coast regions of peninsular India. The aerosol transport from IGP towards south India (referred to as RTE) occurs mainly under the anticyclonic circulation prevalent over the east coast of India and nearby BoB regions. The RTE events typically spans for 2-4 days and are characterized by widespread haziness across the eastern coast. While the pollutant concentrations during RTE is highest among the northern latitudes, it gradually decreased over south. Such aerosol transport has induced occurrence of an aerosol layer, referred as TAL, having thickness of ~1-2 km departed from ABL. While the TAL has relatively lower vertical extension over the northern latitudes (~ 2km), it eventually broadens over the southern latitudes (~3 km). Moreover, occurrence of TAL constituted an atmospheric warming up to ~1-1.5°C where it present. The RTE episodes, in general, promotes the lower tropospheric stability and hence suppress the growth of ABL. Such inhibition of the ABL growth has a latitudinal heterogeneity. Overall, the occurrence of RTE and TAL have suppressed the ABL-H by ~40% and such suppression are dominant during the afternoon hours. Finally, the relation between the ABL-H and $PM_{2.5}$ aggravations are delineated. For instance, a 30% reduction in the ABL-H can contribute to ~130-150% increase in the surface $PM_{2.5}$ concentrations. The diurnal variation of the $PM_{2.5}$ suggests an overall enhancement of ~55% during RTE compared to clear day; however, such enhancements are dominant between 09-11 LT.

This study elucidates the first qualitative investigation of the transboundary transport of aerosols over the Indian peninsula and is a reference for emission policies over the eastern coasts, especially over Chennai and the surrounding area. The analysis of the TAL is carried out by removing the cases of shallow clouds occurring frequently during the study period, which we would like to pursue in a future study.

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Data Availability

MODIS and MERRA2 data can be obtained from NASA Goddard Earth Sciences Data and Information Services Center (GES DISC). CALIPSO data used in this study can be obtained directly from the website https://eosweb.larc.nasa.gov/project/calipso/calipso_table. Radiosonde and surface data can be obtained from the https://weather.uwyo.edu/upperair/sounding.html. The MPL data used in this study are not publicly available; however, the data can be provided to the corresponding author upon request.

Author contributions.

SA was responsible for carrying out the investigation, writing, reviewing, data curation, and preparing the original draft of the paper. CS is responsible for conceptualizing, methodology and supervising, carrying out the investigation, writing, reviewing, and editing the paper. SKM is responsible for MPL data curation, reviewing and editing the paper

Competing interests.

The contact author has declared that neither they nor their co-authors have any competing interests

Acknowledgements

SA thanks the Asia-Pacific Network for Global Change Research (APN) research grant (CRRP2022-08MY-Sarangi) for supporting this work. CS is grateful to MoES Deep Ocean mission project for the support of computational facility for this work.

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The occurrence of RTE days generally prolongs for more than a day. The largest lapsed RTE were observed during March 2022, which continued for 12 consecutive days. The widespread haziness over the northeastern region of the Indian subcontinent induced by the occurrence of TAL, especially during the winter season, is mainly favoured by the relative reduction of wind speed during the season near the surface. The reduced windspeed declines the dispersion of pollutants over a large area in the southern Indian peninsula hence reducing any diurnal variability in the aerosol distribution. On the other hand, although the overall WS declines during RTE days compared to clear days, it shows some enhancement above ~1.5 km, promoting the formation and growth of TAL aloft the ABL. ¶
The TAL has a ~1-2 km thickness and occurs just above the ABL. A strong temperature inversion between the top of the TAL and free troposphere is observed during the RTE periods, followed by a strong warming up to ~1-1.5°C where the TAL is present, constituting an increment of ~5-10% relative to clear sky days. While the TAL has relatively lower vertical extension over the northern latitudes (~ 2km), it eventually broadens over the southern latitudes (~3 km). The RTE episodes, in general, promotes the lower tropospheric stability and hence suppress the growth of ABL. Such inhibition of the ABL growth has a latitudinal variability. Overall, high resolution lidar observations over Chennai suggest that the RTE and occurrence of TAL have suppressed the ABL-H by ~38%; however, for a typical RTE episode, temporal variation in the aerosol extinction characteristic suggests a suppression of up to ~78 %. The occurrence and maintenance of TAL during the RTE are favoured by the strong wind flow from north India, which majorly contributes to the reduction of the ABL-H and aggravation of surface pollution. The aerosol "dome effect", as a result of the vertical temperature stratification due to the presence of TAL, has induced the suppression of the ABL-H and increased the surface $PM_{2.5}$ by ~30-35% compared to the clear days. This study elucidates the first qualitative investigation of the transboundary transport of aerosols over the Indian peninsula and is a reference for emission policies over the eastern coasts, especially over Chennai and the surrounding area. The analysis of the TAL is carried out by removing the cases of shallow clouds occurring frequently during the study period, which we would like to pursue in a future study.¶
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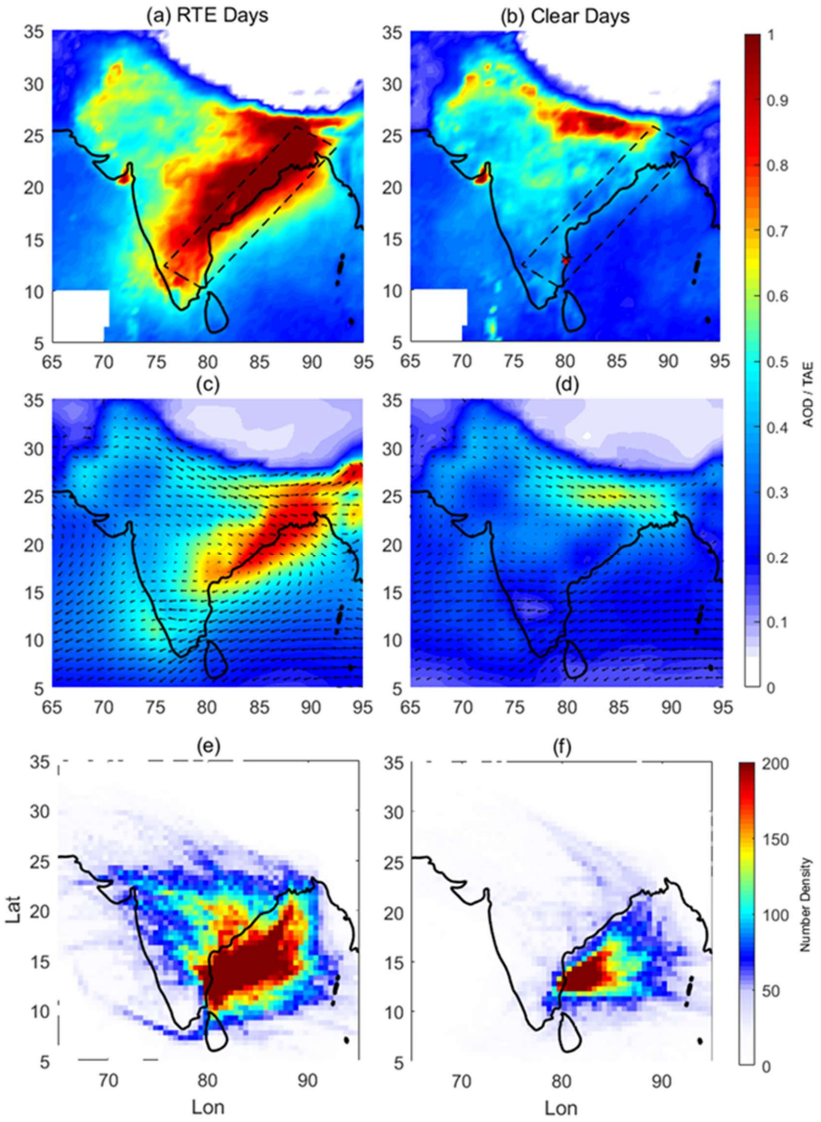

Figure 1 Composite of the spatial distribution of AOD obtained from MODIS during (a) RTE and (b) clear days between December and March during 2015-2024 and total aerosol extinction (TAE) from MERRA2 reanalysis dataset observed for the composite of (c) RTE and (d) clear days. Number density of 5-day backward air mass trajectories to Chennai between surface and 4 km during (e) RTE and (f) Clear days.

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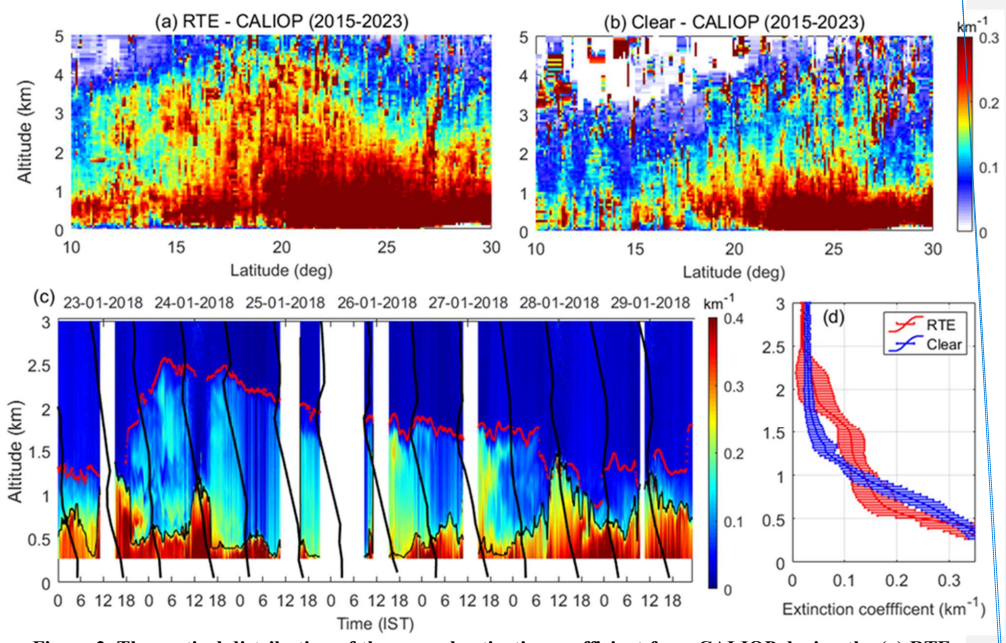
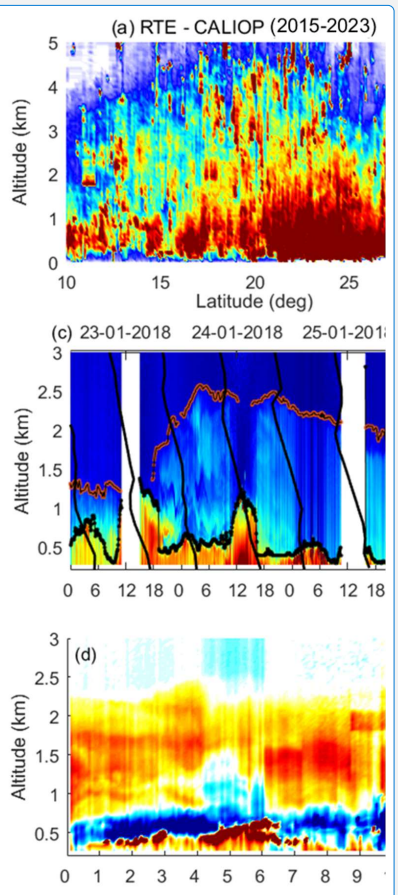


Figure 2. The vertical distribution of the aerosol extinction coefficient from CALIOP during the (a) RTE and (b) clear days within ±5° longitude over the eastern coast of India between December and March 2015-2023. (c) Time-Altitude cross-section of the total attenuated extinction coefficient obtained from Micro Pulse Lidar (MPL) observation over Chennai (SRM IST) between 23 and 29 January 2018. The black line corresponds to the temperature profiles from radiosonde over IMD, Chennai. The black dotted line corresponds to the derived ABL-H, and the red dotted lines are the top of the TAL. (d) Mean extinction coefficient observed during a typical RTE (24- - 27 Jan 2018) and clear day (23,28,29 Jan 2018) estimated from MPL observation.



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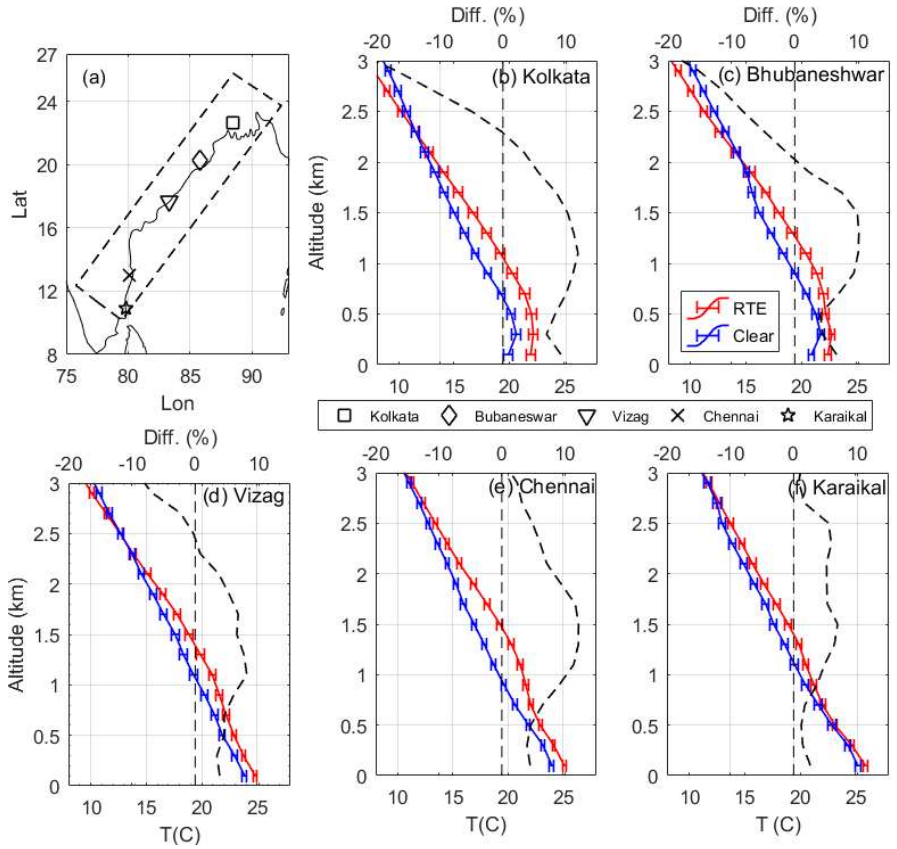
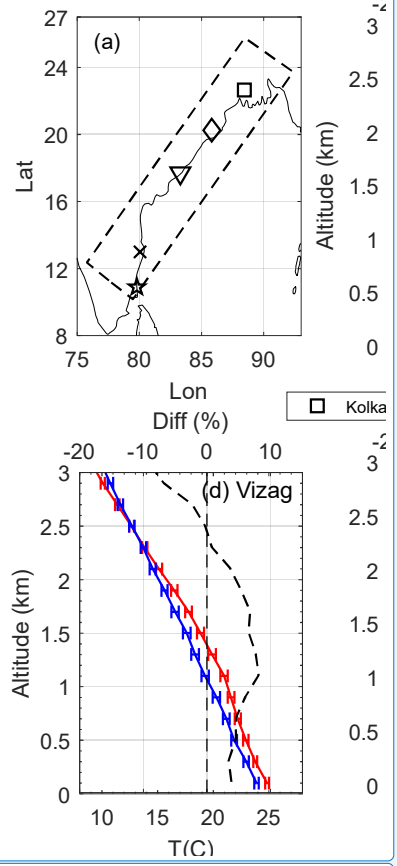


Figure 3 Vertical profiles of temperature during RTE (red) and Clear (blue) days obtained over the different station along the eastern coast. (a) Locations of the Radiosonde observations. (b)-(f) mean temperature profiles with standard errors over the stations Kolkata, Bhubaneshwar, Vizag, Chennai and Karaikal during RTE and Clear days, and the difference between the RTE and Clear in percent (axis on the top). Vertical dashed line corresponds to the 0%.



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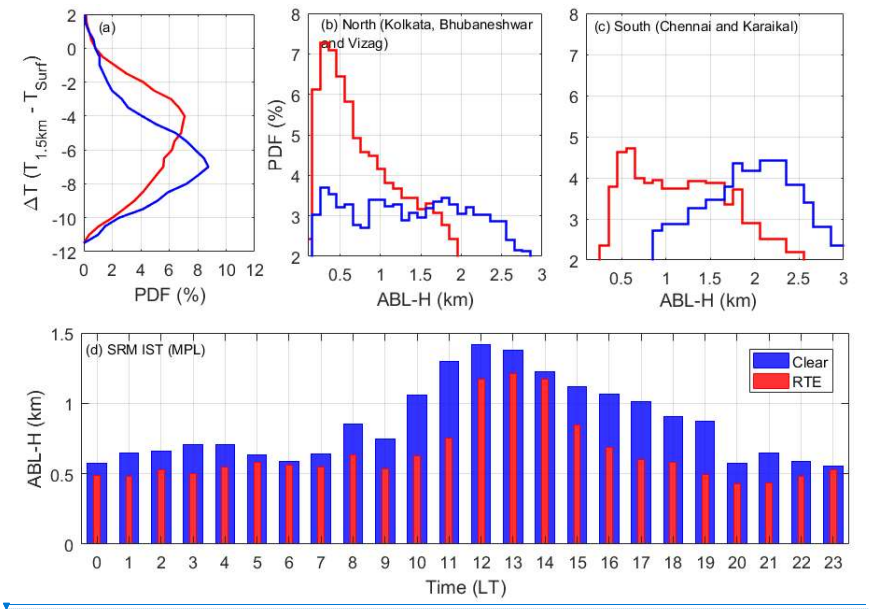

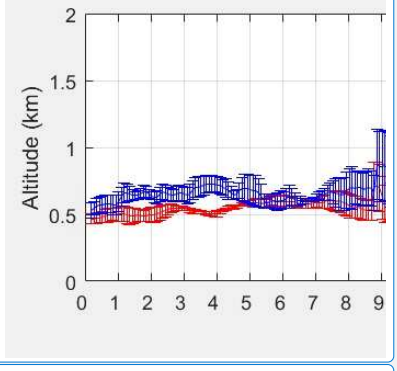

Figure 4. (a) The probability distribution of difference in the temperature between 1.5 km and surface during RTE (red) and clear (blue) days, obtained across all the station over the eastern coast. Probability distribution of ABL-H across (b) the north stations (Kolkata and Bhubaneshwar and Vizag, (c) Chennai and Karaikal obtained during RTE (red) and clear(blue) days. (d) Diurnal variation of the mean ABL-H observed over SRM IST from MPL observation during RTE and clear days.

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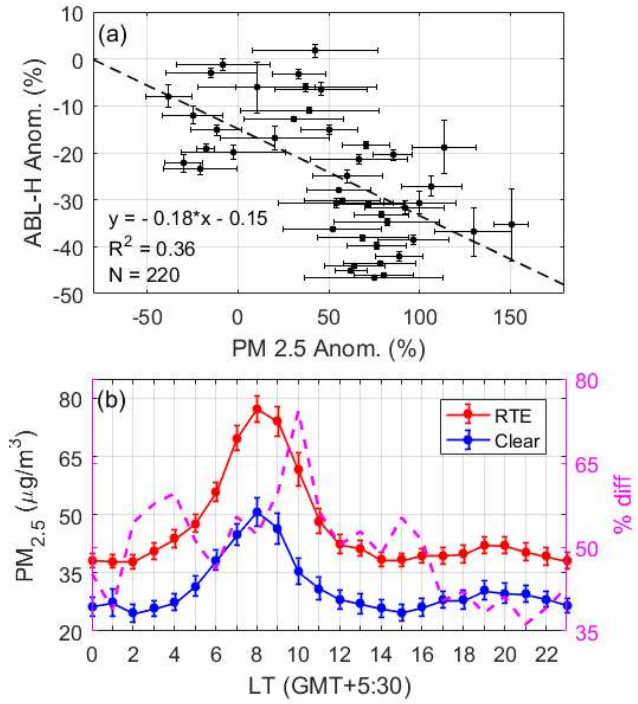
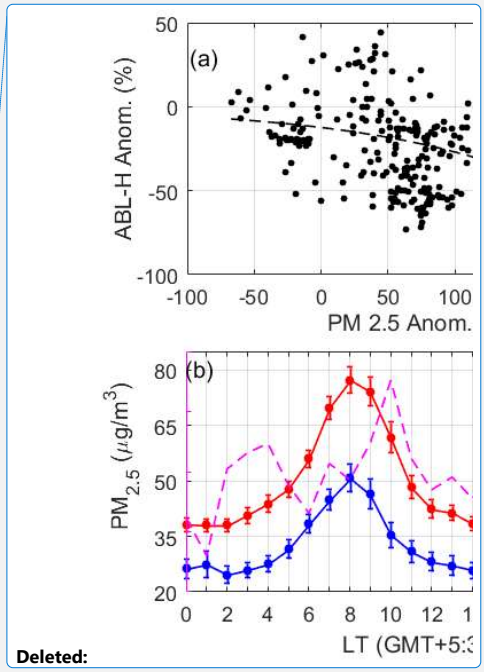


Figure 5. a) Scatter plot showing the normalized anomaly of $PM_{2.5}$ and ABL-H (obtained from MPL, in percent) during the RTE days as observed by MPL. The linear fit, R^2 and number of samples (N) are also provided. b) Diurnal variation of $PM_{2.5}$ over Chennai (US Consulate, Chennai) during RTE and clear days. The percentage difference in the $PM_{2.5}$ (%diff, magenta color) is shown in right axis.



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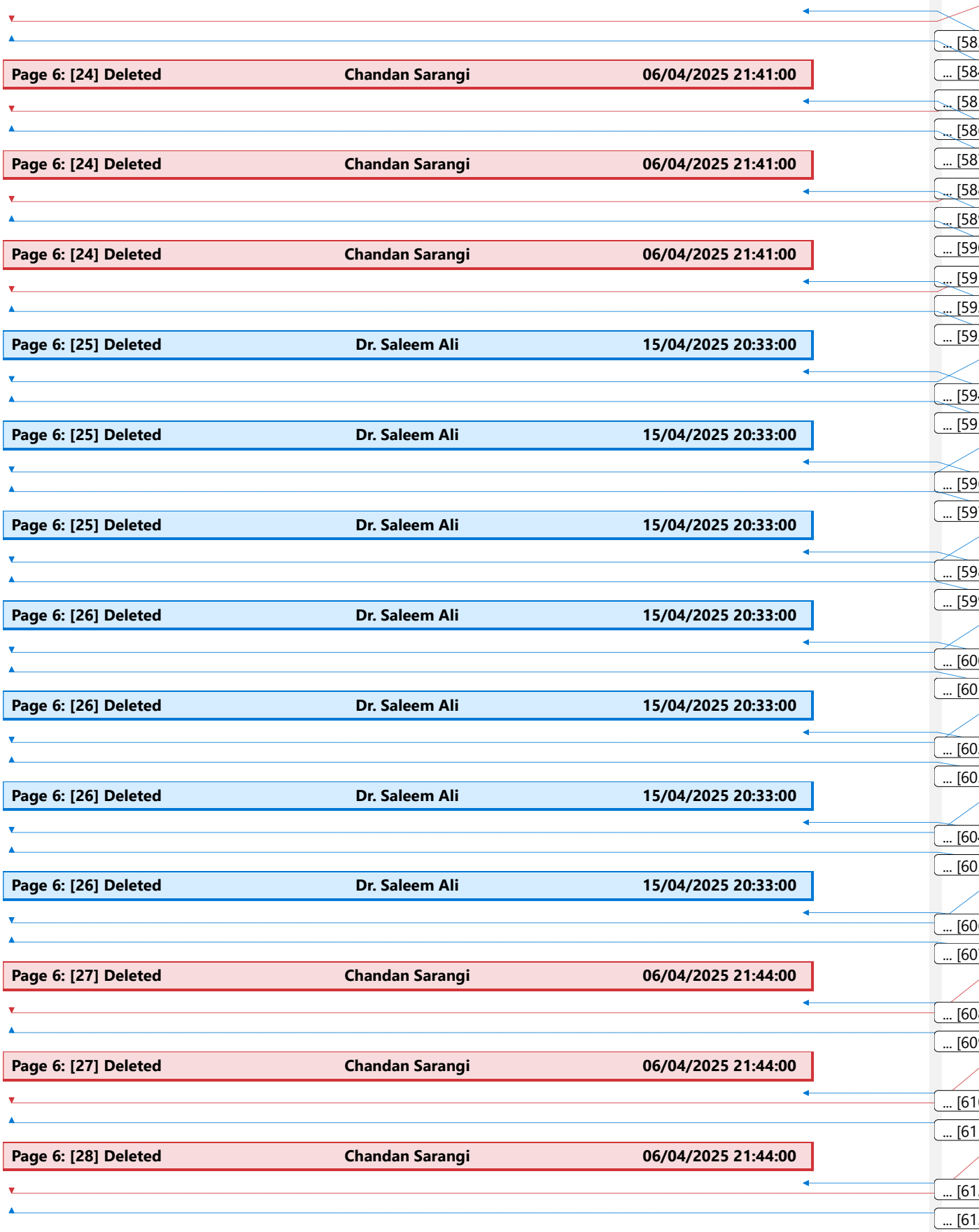
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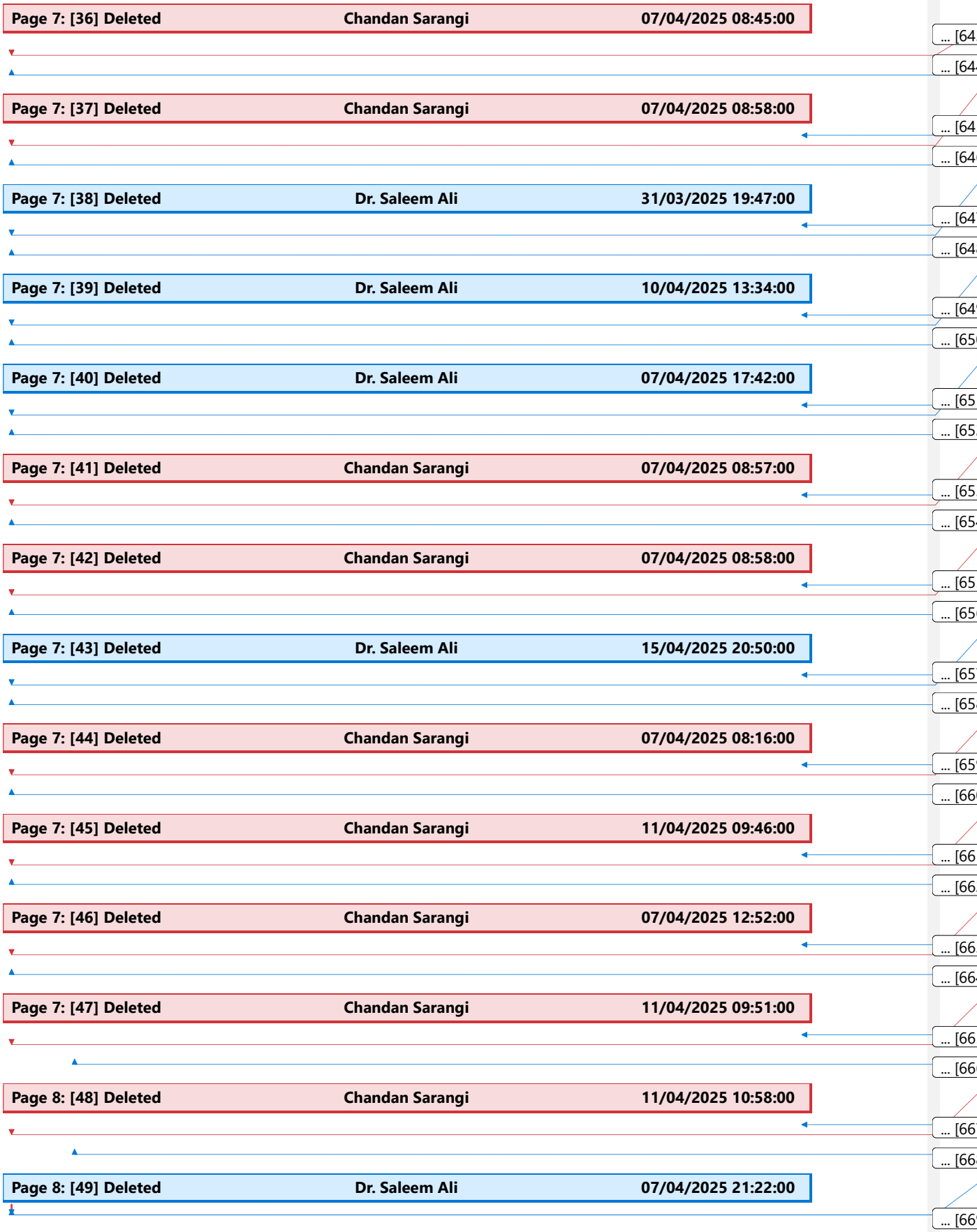
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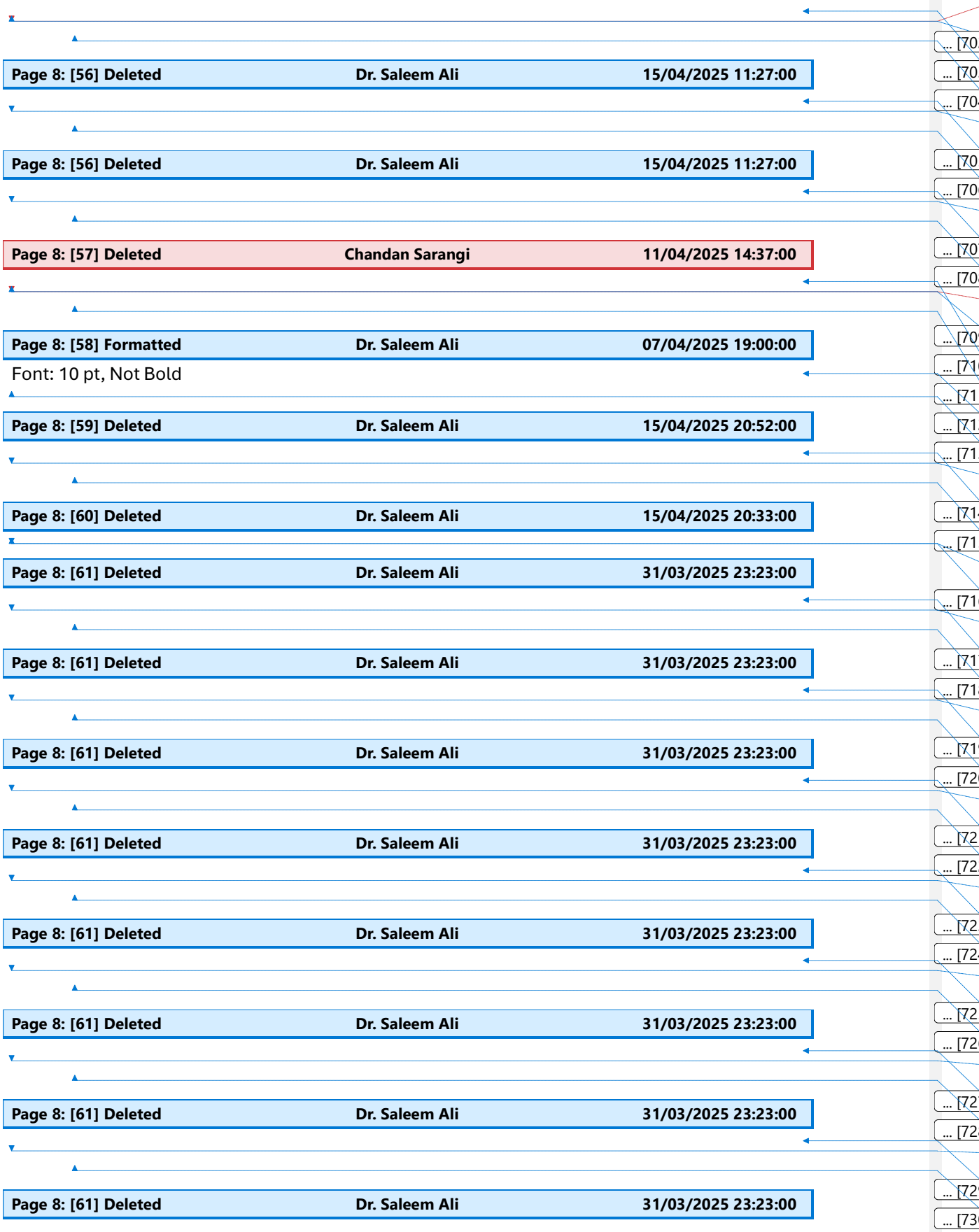
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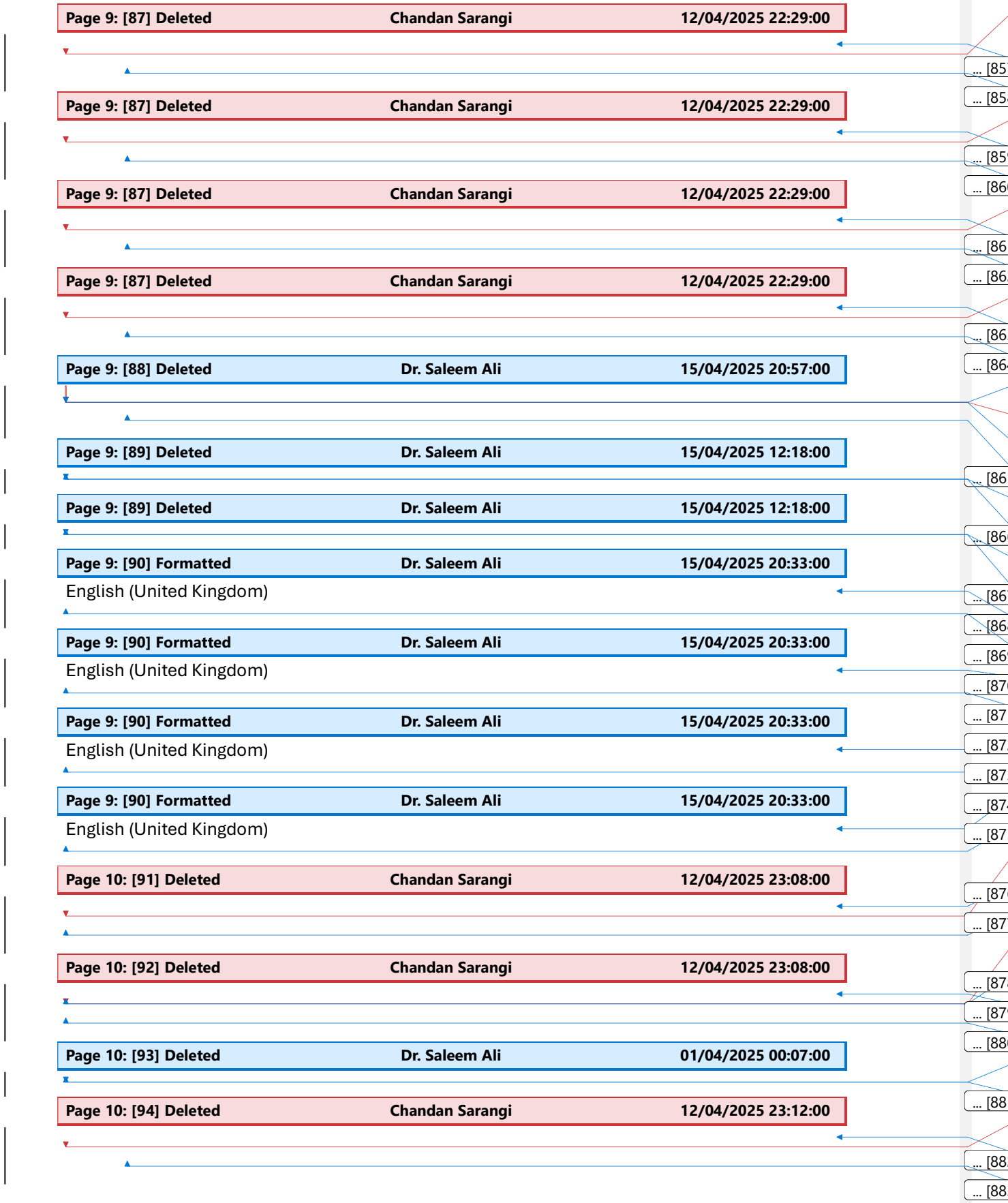
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