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Key Points:

- Aerosol vertical distribution under different static stability regimes is investigated in the Arctic
- As atmospheric stability increases, more and thicker aerosol layers below inversions during winter and above during spring are observed
- For the first time, collocated satellite observations are used to quantify this relationship and to infer the role of long-range transport

Supporting Information:

- Supporting Information S1

Correspondence to:

M. A. Thomas,
manu.thomas@smhi.se

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The Relation Between Aerosol Vertical Distribution and Temperature Inversions in the Arctic in Winter and Spring

Manu Anna Thomas¹ , Abhay Devasthale¹ , Michael Tjernström^{2,3} , and Annica M. L. Ekman^{2,3}

¹Swedish Meteorological and Hydrological Institute, Norrköping, Sweden, ²Department of Meteorology (MISU), Stockholm University, Stockholm, Sweden, ³Bolin Center for Climate Research, Stockholm University, Stockholm, Sweden

Abstract Temperature inversions, being a common meteorological phenomenon in the Arctic, especially during winter and spring, play a special role in regulating the distribution of aerosols. Using 10 years of collocated satellite observations, we evaluate the covariability of the vertical distribution of aerosol extinction and different atmospheric stability regimes. It is observed that as the atmospheric stability increases, more aerosols are trapped below inversions over the majority of the Arctic (except in the Pacific sector) during winter, indicating low-level transport from lower latitudes into the Arctic. In contrast, additional aerosol layers are accumulated above inversion layers during spring in the Eurasian and Pacific sectors as the stability increases, indicating long-range transport at higher altitudes. In this study, satellite observations are, for the first time, used as sole evidence to support the hypothesis that low-level transport dominates in winter and free tropospheric transport dominates in spring.

Plain Language Summary Both in situ measurements and satellite observations unequivocally show that temperature inversions are common phenomena in the Arctic during polar winter. This invisible barrier controls the atmosphere and surface interactions, including aerosol vertical distribution. Having knowledge about the relationship between aerosols and inversions is important as the radiative and dynamic impacts of aerosols in the Arctic are strongly dependent on how they are distributed in the atmosphere in the presence of inversions. However, there exists a key knowledge gap about this relationship. Here we use 10 years of observations from two satellite missions, one providing temperature and humidity profiles and the other providing aerosol vertical distribution, to quantify this relationship for the first time. We investigated aerosol vertical distribution under different stability regimes that are defined based on temperature inversion strength. We found that as the atmosphere becomes more stable, more aerosols are trapped below inversions over the majority of the Arctic (except in the Pacific sector) during winter. During spring, the accumulation of aerosols over the inversion layers is dominant, except in the Atlantic sector. We argue that different pollution pathways into the Arctic can explain the observed contrasting relationships.

1. Introduction

In the 1950s, American and Canadian pilots reported low visibility during early spring over the North American Arctic, referring to it as Arctic haze (Greenaway, 1950; Mitchell, 1957). This phenomenon was later found to be caused by elevated levels of aerosols (Li & Barrie, 1993; Quinn et al., 2002). Temperature inversions, both surface and elevated inversions prominent during winter and spring over the Arctic, efficiently trap aerosols in the boundary layer preventing them from dispersing, hence creating a haze (Hirdman et al., 2010).

Since the first sightings of Arctic haze, more advanced measurements have been performed to understand the sources of Arctic haze aerosols, if they are produced locally or somewhere else afar and transported to the Arctic, and their seasonal variability (Sharma et al., 2004; Tunved et al., 2013). In the recent decade, several Arctic campaigns have examined the vertical distribution and chemical composition of aerosols at several locations in the Arctic. These campaigns were mostly carried out in early spring and summer and reported on long-range transport of pollutants from Eurasian forest fires and Asian anthropogenic sources into the Arctic (Brock et al., 2011; Croft et al., 2016; Curry et al., 2000; Freud et al., 2017; Frossard et al.,

2011; Hirdman et al., 2010; Law et al., 2014; Wang et al., 2011; Warneke et al., 2010). As the summer approaches, the importance of photochemistry and local aerosol production increases. However, surface aerosol concentrations are generally low during summer compared to other seasons, mainly due to efficient wet scavenging and less efficient transport from midlatitudes (di Pierro et al., 2013; Engvall et al., 2008; Garrett et al., 2010; Sharma et al., 2004).

Understanding observations of Arctic pollution demand that we understand the transport pathways by which the pollutants (aerosols and aerosol precursors) reach the Arctic and contribute to aerosol variability (e.g., the Arctic Monitoring and Assessment Program, AMAP, 2015). The transport pathways are often complex (e.g., Stock et al., 2014). In a comprehensive study, Stohl (2006) established the following possible pollutant transport pathways into the Arctic.

1. The most effective and prominent pathway is argued to be direct low-level transport of air masses into the Arctic from lower latitudes. This pathway is more significant during winter as the Arctic front extends farther south over the Eurasian peninsula compared to the summer months, even as far south as 40°N (Barrie, 1986).
2. (a) One other transport pathway from lower latitudes outside the Arctic dome is via low-level transport and subsequent lifting of pollutant-laden air masses at the Arctic dome as the air mass could be warmer, thereby inhibiting low-level isentropic transport. The lifting could subsequently cool the air mass and eventually result in slow sinking or mixing into the Arctic air mass. (b) Another alternative would be the direct injection into the upper troposphere/lower stratosphere at the source itself followed by upper tropospheric transport and then gradual mixing into the Arctic air mass.
3. The third pathway by which the pollutants can be transported to the Arctic is via warm conveyor belts in midlatitude cyclones, transporting the pollutants in the free troposphere into the Arctic.

Pathway #1 is considered to be efficient in transporting pollutants from northern Eurasia during winter and early spring (Hirdman et al., 2010; Sirois & Barrie, 1999; Stohl, 2006). Pathway #2 also transports pollutants during winter and early spring from the northern regions of Eurasia into the Arctic but at higher altitudes (Brock et al., 2011; de Villiers et al., 2010; Frossard et al., 2011; Hirdman et al., 2010; Singh et al., 2010; Stohl, 2006). The third pathway is more relevant for transporting the pollutants from eastern North America during winter.

Once transported into the Arctic, the final fate of the aerosols and their overall impact on the atmosphere and surface depends heavily on local meteorological conditions. This is one of the research areas where comparatively little is known about the Arctic, due to the lack of suitable observations. Temperature inversions, both surface and elevated, are very commonly observed meteorological phenomena in the Arctic especially during winter and early spring (Tjernström & Graversen, 2009). They influence the dispersion of aerosols, their contribution to the vertical distribution of radiative heating, and the way aerosols affect Arctic cloud and radiation processes (Coopman et al., 2016). Characterizing covariability of temperature inversions and aerosol distribution would shed light not only on the transport pathways into the Arctic, depending on if aerosol accumulation occurs above the inversion layer or below, but also help in better understanding aerosol radiative effects. Furthermore, such covariability provides observationally anchored constraints to evaluate the coupling of local meteorology and aerosol distribution in chemistry transport and global climate models.

Various measurement campaigns have so far largely been limited to the summer months, thus not measuring aerosols during those time periods when the transport into the Arctic is most efficient. The launch of the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP)-CALIPSO made it possible to characterize the vertical distribution of aerosol extinctions also in the cold Arctic winter atmosphere (Winker et al., 2013). Therefore, we attempt to characterize this covariability, using simultaneous measurements of aerosol extinctions from CALIOP and temperature profiles from Atmospheric Infrared Sounder (AIRS), both part of NASA's A-Train payloads.

2. Data and Methodology

We use 10 years of AIRS and CALIOP data, from September 2006 to August 2016. Specifically, we use the most recently available Level 2, Version 4.10, 5-km standard aerosol layer product from CALIOP-

CALIPSO to characterize the vertical distribution of aerosol extinction (CALIPSO Science Team, 2016). In the Arctic, so-called diamond dust (wind-blown snow particles, thin ice fog) can sometimes be misclassified as an aerosol layer, although the frequency of occurrence of these cases may be very low (Di Pierro et al., 2013). CALIPSO products also provide information on skills in delineating aerosols and clouds through the Cloud Aerosol Discrimination score. We used Cloud Aerosol Discrimination score ranging from -100 to -50 to delineate aerosol layers from other features. Aerosol layer properties, such as the top and bottom altitude, optical depth, and volume depolarization ratio, are analyzed. One of the inherent limitations of spaceborne lidar is the contamination from the surface signal in the lowermost 100 m (Tackett et al., 2018). This leads to aerosols layers closest to the surface being undetected or discarded causing unrealistic low values of aerosol frequency near the surface. Each aerosol layer detected by CALIOP in the Arctic is mapped onto a $1^\circ \times 1^\circ$ grid to collocate with profiles of temperature and humidity from the AIRS Standard Twice Daily Level 3 product. AIRS data have proven very useful to characterize the vertical thermodynamic structure of the Arctic lower troposphere (e.g., Devasthale et al., 2010; Devasthale, Sedlar, & Tjernström, 2011; Devasthale et al., 2016).

Temperature retrievals at the surface, 1,000, 925, 850, 700, 600, 500, 400, and 300 hPa, are analyzed. A temperature inversion is a layer in the atmosphere where the temperature increases continuously with height. Using the surface temperature as the first reference, a surface inversion thus extends through a layer where the temperature is increasing layer by layer and its top is defined as the highest layer where the temperature is continuously higher than the reference below. If the first level above the reference level has a lower temperature than the surface, the reference temperature is moved up recursively level by level in order to capture elevated inversions. If an elevated inversion is found, the inversion base is defined as the reference level and the inversion top as the level immediately below where the temperature again starts to decrease. The new reference level is then taken as the level immediately above this, and so on to the top of the profile. This procedure delineates between surface and elevated inversions and also allows multiple inversions in the same profile to be detected. The inversion strength is simply the temperature difference between the top and the base of the inversion. The peak inversion height (hereafter, in the text referred to as the inversion top) is the height at which this inversion strength is at its maximum and thus determined instantaneously. In the present analysis, both surface and elevated inversions are considered.

Spatial maps of the climatological median inversion strengths as a function of month are prepared using AIRS data, separately for the ascending and descending passes of the Aqua satellite. If an aerosol layer is detected by CALIOP, the corresponding temperature profile from AIRS for that ascending or descending pass is extracted to analyze the presence of an inversion. If an inversion is detected, its maximum strength is computed and then compared to the climatological median value for that month. Inversions occur in over 80% of all profiles over sea ice during winter and in almost all profiles over multiyear sea ice (supporting information Figure S1). We then ascertain to what degree the inversion strength deviates from climatology. The inversion strength thresholds of 10th, 50th, 70th, and 90th percentiles (hereafter P10, P50, P70, and P90, respectively) are used to define different atmospheric stability regimes to investigate how these regimes relate to the aerosol vertical distribution (Figure 1, left).

We focus on four sectors in the Arctic Ocean (supporting information Figure S2). The selection of these sectors is based on the synthesis of previous studies that not only investigated pathways of long-term pollutant transport into the Arctic (AMAP, 2017, and references therein) but also investigated circulation patterns that favor moisture intrusions into the Arctic (Johansson et al., 2017; Woods et al., 2013), thereby potentially bringing polluted air masses rapidly into the Arctic from lower latitudes. Selected sectors, northward of 70°N , are denoted as AT (Atlantic, 30°W – 25°E), BK (Barents and Kara Seas, 25° – 90°E), PC (Pacific, 145°E – 130°W), and LB (Labrador Sea, 90° – 30°W). A majority of aerosol layers are observed below 1 km, where depending on the sector and season, nearly 3,000 to 4,500 cases are analyzed in the P10 case and at least few hundred cases in the P90 case for each sector. This represents from about 20% of all CALIOP profiles in the P10 case to about 1% of all profiles in the P90 case (cf. supporting information Figure S3).

The vertical distribution of aerosol extinction and optical depth as a function of atmospheric stability regimes are studied and presented in the next section.

Figure 1. (left) Schematic showing analysis methodology adopted in the present study. Atmospheric stability regimes are classified based on four percentile thresholds (10%, 50%, 70%, and 90% denoted as P10, P50, P70, and P90 respectively) applied on the climatological distribution of inversion strengths at each $1^\circ \times 1^\circ$ grid box and for each month separately. The degree of atmospheric stability thus increases from P10 to P90. Higher stable regimes are by definition subsets of the previous regime. (right) The schematic also shows the separation of aerosol vertical distribution below and above the vertical level at which maximum temperature inversion strength is observed. This figure is available in colour online at wileyonlinelibrary.com/journal/grl.

3. Results and Discussion

The vertical distribution of aerosol layers averaged over the winter months December through February under different atmospheric stability regimes for the different sectors is presented in Figure 2 (top and middle rows). The color-coded numbers in each subplot denote the percentage of aerosol extinction layers occurring below the top of the strongest inversion among all the aerosol layers detected in the tropospheric column (up to 10 km); see Figure 1 for an explanation. The vertical distribution is calculated as follows. For each sector and season, we start with four empty height vectors (binned at 250-m vertical bins) representing four stability regimes (i.e., P10, P50, P70, and P90). If an aerosol layer is detected, the presence of an inversion is checked, and if an inversion is present, its strength is compared to climatology to determine under which stability regime the aerosol layer was observed. The mean height of that aerosol layer is then calculated, and the corresponding bin in the height vector in that stability regime is incremented. In the end, each height vector is normalized by the total numbers of aerosol layers that end up in that regime, thus obtaining the normalized vertical distribution of aerosol layers in each regime, sector, and season.

Supporting information Figure S4 shows the histograms of top heights of temperature inversions during different stability regimes. During the majority of the time, the top of the temperature inversion is around 850 hPa (~ 1.5 km) in the P90 case, between 850 and 925 hPa (~ 750 m) in the P70 case, and close to 925 hPa in the P50 and P10 cases.

Irrespective of the sector selected, the aerosol layers are predominantly observed below 1 km during the winter months (Figure 2, top and middle rows), with increased frequency of occurrence in the AT and BK sectors compared to the PC and LB sectors. This climatological vertical distribution of aerosol layers is broadly consistent with previous studies (Devasthale, Tjernstrom, et al., 2011; Di Pierro et al., 2013; Doherty et al., 2010). As the degree of atmospheric stability increases (from P10 to P90), a larger fraction of aerosol layers are detected below the inversion top in the AT, BK, and LB sectors. For example, in the AT sector, 68% of all aerosol layers lie below the inversion top height during extremely stable conditions (P90), compared to 47% during climatological conditions, indicating an increase up to 21% increase with increasing stability. In the BK and LB sectors, the corresponding percentage increases from climatological conditions to extremely stable conditions are 17% and 14%, respectively. This is expected as mixing across the inversion top would be significantly reduced in P90 compared to the P10 case. In contrast, the frequency of occurrence of aerosol layers below inversion top in the PC sector decreases by 15% as the atmospheric stratification increases.

Figure 2. (top and middle rows) Winter (DJF) mean aerosol vertical distribution under different stability regimes for the chosen four sectors. The numbers in each panel indicate the fraction of aerosol layers below inversion in percent. (bottom row) Average aerosol layer optical depths below (left) and above (right) the inversion top height for different sectors and stability regimes. The vertical bars show one standard deviation. DJF = December–February; AT = Atlantic; BK = Barents and Kara Seas; PC = Pacific; LB = Labrador Sea; AOD = aerosol optical depth.

The aerosol optical depths (AODs) above and below the inversion layer under different stability regimes are shown in Figure 2 (bottom row) for the different sectors. The information on both frequency of occurrence of aerosol layers and AOD is needed to fully understand how the opacity of the atmosphere changes above or below the inversion layers. For example, the frequency of occurrence of aerosols can increase without a corresponding increase in total extinction if the aerosol layers are relatively clean. Figure 2 (bottom row) shows a consistent association between AOD and stability in the different sectors. The average AOD below the inversion top steadily increases from P10 to P90 in the AT, BK, and LB sectors, while a decrease is found in the PC sector. The largest increase is observed in the AT sector, where the average AOD below inversion layer increases from 0.084 (P10) to 0.098 (P90). The AOD values above the inversion layers show opposite tendencies, again, except in the PC sector (i.e., a decrease with increasing stability). The frequency of occurrence of aerosol layers above the inversion layers in all the sectors is generally lower compared to those below the inversion layers, but whenever the aerosol layers are detected above the inversions, they are not significantly cleaner.

Aerosol extinction depends not only on aerosol mass concentration but also on the humidity levels and subsequent water uptake. Figure 3 shows the vertical distribution of specific humidity anomalies over the Arctic (70–90°N) for the different sectors, for the P90 case calculated with respect to the 10-year seasonal climatology derived from AIRS during winter and spring. The meridional-height anomalies of specific humidity (g/kg) observed during different stability regimes and averaged over the cases pertaining to all sectors is shown in supporting information Figure S5. It is evident that the humidity levels are increased in the P90 case, indicating additional moisture being transported during these events. Considering the fact that not only the AODs increase (Figure 2, bottom row) but also the frequency of occurrence of aerosol layers (Figure 2, top and middle rows), it is nevertheless likely that additional aerosols are being transported and accumulated as the atmospheric stability increases.

Figure 3. Vertical profiles of specific-humidity anomalies (g/kg) with respect to the 10-year climatology (2007–2016) for P90, derived from Atmospheric Infrared Sounder during winter (left) and spring (right), for the four selected sectors. DJF = December–February; MAM = March–May; PC = Pacific; AT = Atlantic; BK = Barents and Kara Seas; LB = Labrador Sea.

The volume depolarization ratios (dv) as a proxy for the nonsphericity of the particles are investigated. Supporting information Figure S6 shows mean dv values under different stability regimes and below the inversion. It is interesting to note that dv tendencies across the stability regimes roughly correspond to the tendencies in the AODs, indicating that the nonspherical aerosols (such as smoke and black carbon contaminated layers) might be contributing to the increased AODs as the stability increases and aerosols accumulate below inversions. The changes in dv are however not as strong. This is expected considering the fact that, during the transport, aerosols undergo mixing and changes in their morphology. The mean dv values are generally higher (the Probability Distribution Function of dv are much broader) in spring than in winter. Among the sectors, the depolarization ratios over the LB sector are highest.

In boreal spring (March through May), presented in Figure 4 (top and middle rows), the frequency of occurrence of aerosol layers above the inversion layer is overall higher compared to that in the winter months. The fraction of aerosol extinction layers trapped below the inversion top increases with increasing atmospheric stability in the AT sector, but only slightly. In contrast, in the BK and PC sectors the fraction of aerosols below the inversion top generally decreases as the stability increases. No notable difference in the fraction of aerosol extinction layers with varying stability is observed in the LB sector. At the same time, there is an increase in the fraction of aerosol layers above the inversion top with increasing stability in all the sectors. This tendency is most pronounced in the PC sector, leading to nearly 15% increase in the aerosol occurrence aloft as the atmosphere stability increases from P10 to P90. In the BK sector, this increase amounts to 8%. The average AOD values during boreal spring above the inversion layer (Figure 4, bottom row) also show consistent increase as the stability increases in the PC, BK, and LB sectors. For example, the average AOD above the inversion top increases from 0.056 to 0.080 in the PC sector and from 0.059 to 0.076 in the BK sector. It has to be noted that even though the frequency of occurrence of aerosol layers below the inversion layer is much lower in spring compared to the winter months, the spring AODs are much higher, which means that the aerosol layers are comparatively more polluted than in the winter months.

How can the covariability of aerosol vertical distribution and inversions in winter and spring seasons be explained? One way is to consider the different transport pathways, as discussed in section 1. Low-level transport within the Arctic dome in winter constitutes one of the major pathways of pollution transport into the Arctic; that is, pathway #1 could explain increasing aerosol accumulation below inversions with increasing inversion strength in the AT, BK, and LB sectors. This could also be a major pathway in the PC sector, although aerosol layers are observed only 29% of the time below the inversion layer. However, the AODs for the PC sector are relatively high, which implies that although the frequency of observing aerosol layers below the inversion is less compared to that in the other sectors, the layers are more polluted. The transport above the inversion top in the PC sector by pathway #2 appears instead to be dominating. Major contributors to the aerosol concentrations in the AT and BK sectors are primarily Eurasian pollutants transported at low

Figure 4. Same as in Figure 2, but for spring (MAM). MAM = March–May.

altitudes in winter (Barrie, 1986; Eckhardt et al., 2003). Emissions from flaring of natural gas by the oil industry in northern Russia is a main source of black carbon (Stohl et al., 2013), and the smelting activities in Norilsk are a significant source of sulfate in the BK sector during this season (Hirdman et al., 2010). The southward extension of the Arctic dome facilitates pollutants from Europe reaching the LB sector.

When Arctic surface starts to warm in summer, the extent of the polar dome decreases reducing low-level injections of pollutants into the Arctic from lower latitudes. Irrespective of sector, however, a different transport pathway, in addition to pathway #1, is common during spring. This involves lifting of air or elevated injection and rapid transport into the Arctic free troposphere, resulting in accumulation of aerosols in the free troposphere. A study by Johansson et al. (2017) showed that a low-pressure anomaly over east Siberia and high-pressure anomaly over Alaska and Beaufort Sea during events of increased moisture transport into the Arctic through the LB sector could favor elevated transport of aerosols over inversion layers. Stably stratified conditions trap the pollutants, preventing them from dispersing. Hence, pathway #2 likely facilitates the transport of pollutants into the Arctic in these sectors. The major contribution of aerosols to the PC sector is from Southeast Asian emissions. Increased frequency of aerosol extinction layers that are observed above inversions in the BK and PC sectors may be as a result of elevated transport of biomass burning aerosols associated with agricultural fires in Eurasia (Brock et al., 2011; McNaughton et al., 2011; Warneke et al., 2009, 2010). Apart from the anthropogenic sources, natural aerosols, mainly sea salt, can also contribute to the observed aerosol variability over the AT and PC sectors; however, their contribution is likely to be limited.

The signature of these transport pathways is also visible in the specific humidity anomalies (Figures 3 and S5). Increased humidity with stronger inversions in winter in all the sectors indicating increased moisture transport can be observed in the lower levels up to 850 hPa indicating low-level transport and, hence, accumulation of aerosols below the inversion top. Furthermore, specific humidity anomalies are twice as high in spring compared to winter in all sectors. The presence of elevated humidity anomalies clearly signals transport via stronger and elevated systems consistent with accumulation of aerosols above the inversion layer in all the sectors, except in the AT sector. These elevated systems seem to be more prominent in the PC sector in spring.

It must be acknowledged that the sources within the Arctic could also have an impact on the statistics presented here. The recent results from the Arctic Climate Change, Economy and Society aircraft campaign (2011–2015) over the European Arctic have provided insights into the local Arctic pollution sources (Law et al., 2017). These sources arise from shipping and oil/gas extraction activities. Though the shipping is the main local source of SO_x and NO_x during spring and summer, the petroleum-related emissions occur throughout the year. Other local sources such as wood burning and metal smelting in winter are also another source of emissions in the Arctic. It is to be noted that these sources remain close to the continental landmass and under favorable circulation patterns these pollutants will be readily transported over the central, permanently sea ice covered areas of the Arctic. These sources fall well within the Arctic dome during both winter and spring, thus contributing to the transport pathway #1 in the AT and BK sectors.

4. Conclusions

In this study we investigate the covariability of aerosol profiles and inversions during winter and spring to explain the distribution of aerosols in the different sectors of the Arctic under different stability regimes and the role of potential transport pathways taken by pollutants to reach the Arctic under these regimes. Ten years of aerosol retrievals from CALIOP-CALIPSO and temperature and humidity profiles from AIRS-Aqua, both sensors being part of the A-Train constellation, allow us to quantify the aerosol vertical distribution during different atmospheric stability regimes. To our knowledge, this is the first comprehensive attempt to shed light on the covariability of meteorology and aerosol vertical distribution during the cold dark months of the Arctic using observations only. The observations show that the fraction of aerosol extinction layers trapped below the inversion top is much higher during winter months compared to spring, supporting the hypothesis that low-level transport from midlatitudes into the Arctic is the most important transport pathway during December through February. The fraction of aerosols trapped below inversion layers during winter also increases as the degree of atmospheric stability increases by up to 21%, 17%, and 14% compared to the climatology in the AT, BK, and LB sectors, respectively. In contrast, more aerosol layers are located above the inversion top during spring months. Moreover, the fraction of aerosol layers aloft also increases with increasing stability by up to 15% in the Eurasian and Pacific sectors supporting the hypothesis that aerosols are lifted before they are transported into the Arctic during March through May. AODs show a consistent increase with increasing stability and with increasing fraction of aerosol layers below the inversion top in winter in the AT, BK, and LB sectors. However, in spring, though the frequency of occurrence of aerosol layers below the inversion layer is lower compared to the winter months, the spring AODs are much higher, which implies that the aerosol layers are comparatively more polluted than in the winter months. Our study provides observational constraints to support process-oriented evaluation of chemistry transport and climate models.

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