RESEARCH ARTICLE

Observation of carbonaceous aerosols during 2006–2009 in Nyainqêntanglha Mountains and the implications for glaciers

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Abstract Atmospheric carbonaceous aerosols were sampled discontinuously from July 2006 to December 2009 at Nam Co Comprehensive Observation and Research Station (NCOS) in the central Tibetan Plateau (TP). The mean daily concentration of carbonaceous aerosols increased from 268 to 330 ngm⁻³, and pollution episodes could significantly increase the mean level of carbonaceous aerosols in the total mass concentration. Organic carbon was the main component of carbonaceous aerosols at NCOS, and black carbon (BC) accounted for 5.8 %. Seven-day air masses backward trajectories calculated by the Hybrid Single-Particle Lagrangian Integrated Trajectory model and the aerosol optical depth distribution in the TP and South Asia both suggested that atmospheric pollutants emitted from Northern India and South Asia could penetrate into central TP by southwest winds. Due to the seasonal variations of emission sources and regional atmospheric conditions, calculated BC deposition flux in the nonmonsoon season was higher than that in the monsoon season. Increased BC

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concentration in snowpack in winter from 2007 to 2009 indicated that the atmospheric environment in central TP became more polluted and the influences from human activities have strengthened. Pollution episodes could significantly increase BC concentrations in the snowpack on a seasonal scale, which would furthermore affect the surface albedo.

Keywords Black carbon · Organic carbon · Snowpack · Pollution · Tibetan Plateau (TP)

Introduction

Carbonaceous aerosols are mainly generated from incomplete combustion of fossil fuels, biofuels, and biomass burning and are composed of organic carbon (OC) and black carbon (BC). OC mainly acts as a light-scattering aerosol to scatter solar radiation, reducing the energy reaching the Earth's surface. It also partly absorbs ultraviolet radiation and shows slight absorption at the visible spectrum, influencing the radiation budget of the climate system (Kirchstetter and Novakov 2004). BC primarily absorbs solar radiation spanning from ultraviolet to near-infrared wavelengths, reducing significantly the spectral albedo of visible light (Warren and Wiscombe 1980). BC suspended in the atmosphere can heat the atmosphere while it cools the Earth's surface by reducing the surface-incident solar radiation. It can also decrease the surface reflectance when it deposits on high-albedo surfaces such as snow or ice (Flanner et al. 2009). By changing the Earth's radiation budget, carbonaceous aerosols can affect clouds, temperature, and precipitation on a global/regional scale.

Carbonaceous aerosols can affect the formation and radiative properties of clouds by acting as cloud condensation nuclei (CCN). The simulation by Spracklen et al. (2011) reveals that carbonaceous aerosols contribute 52-64 % to the global mean surface CCN concentrations at 0.2 % supersaturation, and the proportion is even larger over polluted continental regions. BC has been determined to be an important component of atmospheric brown clouds, exerting significant effects on the Indian monsoon and precipitation pattern in South Asia (Ramanathan and Carmichael 2008). BC emissions from continental regions near the equator simulated by the BC aerosol-climate model could significantly affect tropical convective precipitation (Wang 2009). Bauer et al. (2010) proposed that the net radiative forcing of carbonaceous aerosols depends on their size distribution, shape, and mixing state. According to Jones et al. (2011), BC emitted by fuel combustion increased the global mean near-surface temperature by 0.14 ± 0.1 °C, producing the radiative forcing of +0.25 Wm⁻² over the twentieth century. However, uncertainties induced by carbonaceous aerosols are very large and the net radiative forcing is still uncertain.

In recent decades, BC has always been a concern for the research on climate change due to its strong absorption in the visible spectrum (Warren and Wiscombe 1980). Once BC deposits onto the surface of snow and ice, it can decrease the snow-ice surface albedo, increase surfaceincident radiation, and advance the melting season which accelerates the melting. The albedo of mountain glaciers decreased significantly when they were strongly contaminated by BC (Ming et al. 2009). In our present understanding, BC is considered to be a potential factor that might have affected the observed glacial changes in the Himalayas (Ming et al. 2008; Xu et al. 2009). Estimated BC in surface snow of a Himalayan glacier by Yasunari et al. (2010) could be responsible for 2.0-5.2 % surface albedo reduction in the premonsoon season, and if the albedo reduction continued throughout the year, an additional 70-204 mm of water equivalent runoff would occur in a typical Tibetan glacier. Increasing surface-incident radiation by BC in the snowpack could increase the global mean 2-m air temperature by 0.10-0.15 °C (Flanner et al. 2007).

The Tibetan Plateau (TP) in Southwestern China is a key region where many mountain glaciers develop. These mountain glaciers release melting water and provide fresh water to the lower residents. However, with regional economic development, the environment and climate in the TP are gradually changing. Therefore, measurements of atmospheric carbonaceous aerosols are of great importance to comprehend these changes.

In this study, we focused on in situ measurements of carbonaceous aerosols (OC and BC) at Nam Co Comprehensive Observation and Research Station (NCOS) in central TP. The data spanning from July 2006 to January 2007 has been published by Ming et al. (2010). Here, we will primarily analyze the variation characteristics of carbonaceous aerosols and estimate the lower limit of BC concentration deposited on the glaciers nearby.

Field experiments

Site description and weather conditions

The NCOS (30.77° N, 90.99° E; 4,730 m a.s.l.) in the north of the Nyainqêntanglha Mountains in central TP is approximately 48 km to the closest downtown Dangxiong city. Nam Co Lake covers an area of 1,980 km² with an elevation of 4,718 m (Fig. 1) and NCOS was built at the southeast coast of the lake in 2005. The geomorphology of the surrounding consists of grassland, lakes, high mountains, and glaciers. The surrounding area of the station is sparsely



Fig. 1 Location map of NCOS and the surrounding geographical environment

populated due to the hostile environment and harsh climate. The local inhabitants mainly make a living by grazing and produce very limited atmospheric pollutants. Therefore, NCOS is a remote and pristine site that is relatively far away from industrial and populated areas.

Carbonaceous aerosols at NCOS were mainly affected by the prevailing winds and surrounding emission sources. Meteorological conditions at NCOS show very distinctive seasonal variation. Monsoon prevails from June to October when more precipitation was observed, while westerly winds dominate in the nonmonsoon season (November– May) with less precipitation. During the sampling period of carbonaceous aerosols, SSE winds prevailed during the monsoon season, while WSW winds prevailed during the nonmonsoon season (Fig. 2). It was worth noting that SSW winds were also very strong in the monsoon seasons of 2009, accounting for 36.6 % of total wind directions, secondary to SSE winds (39.2 %) (Fig. 2c).

Sampling and methods

The portable air sampler (MiniVol, AirmetricsTM, USA) used to sample total suspended particulates (TSP) was installed on the roof of the sampling cabin, about 3 m above ground level (AGL). The pump was operated at a stable airflow rate of 5 Lmin⁻¹ which was recommended by the manufacturer with a sampling time resolution of 4–14 days. The volume of each sample was in the range of 14.27–58.41 m³ (at 0 °C and 1,013 hPa). The quartz filters (Whatman[®] QMA, UK) with the diameter of 47 mm were preheated in an oven at 600 °C for 24 h to eliminate any



Fig. 2 The distribution of wind direction frequency at NCOS in the monsoon and nonmonsoon seasons from July 2006 to October 2009: **a** July 2006 to May 2007; **b** June 2007 to May 2008; **c** June 2008 to October 2009

carbonaceous matter and then enclosed in the Petri slides. A quartz filter was mounted in the sampler while sampling and a special cap designed by the manufacturer was set on top of the filter holder to avoid the interference of different weather conditions. Finally, quartz filters loaded with TSP were stored at -20 °C until laboratory analysis. In total, 122 samples were collected at NCOS from July 2006 to December 2009, wherein scarce samples were obtained in the summers of 2007 and 2008 and April and May 2009 because the sampler was out of order.

In the laboratory, 0.5 cm² quartz filter loaded with TSP was punched from the original filter (11.95 cm²) and analyzed by a Desert Research Institute Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., USA). OC and BC were distinguished based on a thermal/optical reflectance method followed by the Interagency Monitoring of Protected Visual Environments protocol. The accuracy of measurements kept within ± 10 %, with a detection limit of 0.2 µg Ccm⁻² (Chow and Watson 2002). The methods to determine OC and BC concentration and detailed heating procedures can be found in the previously mentioned literatures.

Results and discussion

Characteristics of atmospheric carbonaceous aerosols

Period variation and sources of atmospheric carbonaceous aerosols

We divided the data into four periods because of the discontinuous sampling and then analyzed the variation characteristics during every period. The mean daily concentration of carbonaceous aerosols at NCOS was 268 ngm⁻³ during the first period (July 2006-April 2007) and increased to 330 ng m^{-3} during the fourth period (July 2009–December 2009), increasing by 23.1 % (Fig. 3c). The maximum mean daily concentration (340 ngm⁻³) occurred during the third period (October 2008-March 2009). The peak of carbonaceous aerosols occurring in March 2009 was caused by an episode. Xia et al. (2011) confirmed that huge amounts of fresh emission in South Asia were penetrating the TP during that period and led to the high concentration of carbonaceous aerosols. Therefore, under the significant contribution of the episode to the mean daily concentration of the third period, carbonaceous aerosols at NCOS showed an obvious increase from the first period to the third period. Afterwards, it decreased slightly during the fourth period.

Mean daily OC concentration showed almost the same variation pattern as that of the total carbonaceous (TC) aerosols before March 2009 (Fig. 3b), contributing to over 90 % to carbonaceous aerosols, and peaks marked in

Fig. 3 Concentration variations of carbonaceous aerosols (TC, including BC and OC) from July 2006 to December 2009, each step started and ended at the sampling beginning and ending date, and *horizontal lines* for the mean level and *dashed lines* for the mean level while ignoring the peaks



Fig. 3b, c also demonstrated that OC was the primary component of carbonaceous aerosols. Comparable OC proportion of carbonaceous aerosols was also observed at Manora Peak (~82 %), a high-altitude site in North India, which implied primary OC from biomass burning associated with anthropogenic species (Rengarajan et al. 2007). However, the consistent variation between TC and OC began to change during the fourth period and the inconsistency became especially obvious when the peak in October 2009 occurred (Fig. 3c).

The mean level of daily BC concentration had a weak increase from the first period to the third period, with an increasing rate of 1 ng m⁻³ per period. However, BC mean concentration had a dramatic increase since July 2009, and the mean level changed from 13 ngm⁻³ during the third period to 45 ngm⁻³ during the fourth period. Due to weak local emission at NCOS, the BC peak in October 2009 might be associated with another episode. If we ignored the peak caused by the episode which cannot represent the background concentration, the measured mean BC concentration during the fourth period would be 24 ngm⁻³. Figure 3a, c indicates that the TC peak is mainly caused by high BC concentration, which accounted for 54.6 % of carbonaceous aerosols.

We calculated seven-day air masses backward trajectories for the four peaks occurring in Fig. 3 with the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model provided by the Air Resources Laboratory of National Oceanic and Atmospheric Administration, driven with $2.5 \times$ 2.5° gridded reanalysis meteorological data provided by Climate Diagnostics Center (CDC-1) National Centers for Environmental Prediction/National Center for Atmospheric Research. The trajectories were calculated at three different heights (500, 1,000, and 1,500 m AGL) starting at 00:00 Coordinated Universal Time on each day that a peak value occurred. The trajectories show that, once strong regional/local emissions occurs in South Asia and Northern India, emitted pollutants could rapidly penetrate into central TP via atmospheric circulation (Fig. 4). Figure 4a, c indicates that emissions in Northern India could directly invade into central TP by WSW winds during the nonmonsoon seasons. Figure 4b, d indicates that emissions in Northern India could transport along the southern slope of the Himalayas and then penetrate into central TP by southwest winds. Emissions at the southern slope of the Himalayas can directly penetrate into central TP by southwest winds.

Due to the fact that vehicular exhausts contribute more to BC concentrations at urban cities, the OC/BC ratio at NCOS was much higher than the mean ratio of 2.39 ± 0.47 reported by Novakov et al. (2005) at urban sites in China (Fig. 5). Zhang et al. (2010) observed high OC/BC ratios that varied in the range of 2.5-35.0 at background sites in the south and southeast China. The OC/BC ratio around 4.0 is representative of combustions of fossil fuel (Koch 2001), and the OC/BC ratio around 8.0 represents biomass burning emissions (Novakov et al. 2005). High OC/BC ratio at NCOS indicated that OC was the main component of carbonaceous aerosols, suggesting that biomass burning was the primary source. The high BC concentration since July 2009 led to a decrease of the mean OC/BC ratio to 12.6, which meant that the contribution of BC concentration for OC/BC ratio became more apparent during the particular period.

Fig. 4 Seven-day air masses backward trajectories for four peaks occurring in Fig. 3: a 31 December 2006, b 20 January 2008, c 8 March 2009, and d 18 October 2009 (http://ready.arl.noaa.gov/HYSPLIT traj.php)

Compared to BC measurement at other sites all over the world since 2000 (except Waliguan, 1991–1995), the mean

BC concentration at NCOS was relatively low (Fig. 6). Most of the samples at NCOS show BC concentrations below

Fig. 5 Variation of atmospheric OC/BC ratios at NCOS

150 ngm^{-3} . However, BC concentration in the TP is higher than that in the Arctic or Antarctic (Fig. 6; Table 1), which indicates that the TP was more exposed to anthropogenic activities.

Seasonal variation of atmospheric carbonaceous aerosols

Carbonaceous aerosol concentrations at NCOS were higher in the nonmonsoon season than in the monsoon season (Fig. 7i–l); however, the seasonal difference was not too obvious (<100 ngm⁻³). The comparable mean level of TC in the monsoon and nonmonsoon seasons in Fig. 7l is the result of a significant increase in BC concentration during the monsoon season of 2009. Seasonal variation of OC concentration is almost the same as that of TC, and the annual difference of OC seasonal variation is very stable (Fig. 7e– h). Seasonal variation of BC concentration is irregular (Fig. 7a–d). The concentration was higher in the monsoon seasons in 2006 and 2009, while the opposite occurred in 2007 and 2008. Mean levels of BC concentration in the

Table 1 Distribution of BC concentration at remote sites all over the world since 2000

Site	Longitude	Latitude	BC (ngm ⁻³)	Site type	Period	Literature
NCOS	90.99	30.77	127	Remote continental	2006.7-2009.12	This work
Qilian Mountain	96.51	39.5	62	Remote continental	2009.7-2011.3	Zhao et al. (2012)
NCO-P	86.82	27.95	160.5	Remote continental	2006.3-2008.2	Marinoni et al. (2010)
Waliguan ^a	100.9	36.28	135	Remote continental	1991-1995	Wen et al. (2001)
Hanle Valley	78.96	32.78	77	Remote continental	2009.8-2010.7	Babu et al. (2011)
Barrow Zeppelin	-156.6 11.9	71.3 78.9	36 27	Arctic Arctic	2001–2008 2002–2008	Skeie et al. (2011)
Southern Great Plains	-97.5	36.6	329	Continental	2001-2008	
Trinidad Head	-124.1	41.1	163	Marine	2002-2008	
Mauna Loa	-155.6	19.5	17	Marine	2001-2008	
Bondville	-88.4	40.05	493	Continental	2001-2008	
Birkenes	8.25	58.4	121	Continental	2001-2007	
Melpitz	12.9	51.5	1,500	Continental	2004-2007	
Montseny	2.35	41.76	213	Continental	2002-2008	
Rishiri Sado	141.2 138.4	45.12 38.25	360 430	Remote island Remote island	2001.3 2001.4	Ramachandran and Rajesh (2007)
Hachijo	139.75	33.15	330	Remote island	2001.3-4	
Chichi-jima	142.22	27.07	210	Remote island	2001.3-4	
Amami Oshima	129.7	28.44	630	Remote island	2001.4	
Manora Peak	79.5	29.4	1,360	Continental	2004.12	Pant et al. (2006)
Besisahar	84.375	28.232	1,100	Continental	2009.5-6	Shrestha et al. (2010)
Maitri	12	-70	75	Antarctic	Summer in 2009	Chaubey et al. (2010)
Larsemann Hills	77	-69	13	Antarctic	Summer in 2009	Chaubey et al. (2010)
Syowa station	39	-69	88	Antarctic	2004.2-2007.1	Hara et al. (2008)

^a The sampling period at Waliguan site from 1991 to 1995

Fig. 7 Seasonal variation of carbonaceous aerosols (TC, including BC and OC) from July 2006 to December 2009; *M* representing the monsoon season and *N*-*M* representing the nonmonsoon season. Each

monsoon and nonmonsoon seasons both have reached the maximum in 2009, which indicates the change of atmospheric environment in the TP.

The high concentration of carbonaceous aerosols in nonmonsoon seasons was related to emissions and atmospheric transport. Central TP was under the control of WSW winds in the nonmonsoon seasons when biofuel and biomass burning were widely used in South Asia. Meanwhile, the atmospheric condition was stable and adverse to atmospheric diffusion. Therefore, emissions in the southwest of TP could be transported over and change the atmospheric environment. Ramanathan et al. (2007) pointed out that the South Asian haze, spreading northeastward along the south side of the Himalayas under stable atmospheric conditions, persisted in winter and the haze layer could reach 3-5 km in the troposphere. Therefore, the high concentration of carbonaceous aerosols in the nonmonsoon season during the sampling period was likely to be associated with the South Asian haze because pollutants (OC, BC, sulfate, and fly ash) from biomass burning and industrial emissions appeared to be the major components of the Asian brown clouds (Ramanathan et al. 2005).

step started and ended at the sampling beginning and ending date, and *horizontal lines* for the mean level and *dashed lines* for the mean level while ignoring the peaks

Relationship between carbonaceous aerosols and aerosol optical depth

Generally, the variation of aerosol optical depth (AOD) was insignificant during the period of carbonaceous aerosol sampling at NCOS. However, AOD had a dramatic increase and reached the maximum when the maximum concentration of carbonaceous aerosols occurred in mid-March 2009 (Fig. 8a, b). AOD showed an obvious increase before 16 March and decreased to the normal level as was observed during early March. Xia et al. (2011) related the high concentration to an episode, revealing the contribution of South Asian emission sources to short-term atmospheric environment variations and to the changes in carbonaceous aerosol concentrations in the TP. Figure 8c shows that AODs measured by multiangle imaging spectroradiometer (MISR) also show corresponding high peaks with carbonaceous aerosol concentrations in mid-March and mid-October 2009. AODs measured by Aerosol Robotic Network and MISR both demonstrated that emissions from South Asia could be transported into the central TP via atmospheric circulation to affect the whole troposphere in the TP.

Fig. 8 Covariation between carbonaceous aerosols concentration and AOD at 500 nm at NCOS: a green steps for concentration of carbonaceous aerosols and blue circles for AOD measured at NCOS, b blue bars for the variation of AOD and dark solid line for three times polynomial fitting in March 2009. and c blue circles for AOD measured by MISR while sampling of carbonaceous aerosols was lacking and red circles for AOD while peaks of carbonaceous aerosols occurred in March and October of 2009

Compared to the area-averaged AOD at 555 nm obtained by MISR before 10 March 2009, the mean level of AOD increased by 26.7 % (Fig. 9a). The maximum AOD can be found at the south slope of the Himalayas, indicating the existence of heavy aerosol loading in that region (Fig. 9b, c). Increased AOD all over the Tibet during the episode suggests an invasion of pollutants from South Asia through WSW and SSW winds. Distinct differences between the AOD in TP and in South Asia demonstrated that the TP was much cleaner than its surrounding areas. However, large-scale pollutant emissions from South Asia could reach the hinterland of the TP and affect regional atmospheric conditions by long-distance transport.

Estimation of carbonaceous aerosols depositing flux and concentration in the glacial surface snow layer

Assuming that carbonaceous aerosols are uniformly distributed in the atmosphere and deposit continuously onto the surface, the TC deposition flux can be calculated as: TC

AOD distribution for **b** 1 March to 10 March in 2009 and **c** 11 March to 31 March in 2009. The *ellipses* in **b** and **c** represent the area with heavy loading of aerosols in the atmosphere

deposition flux = TC concentration(ng m^{-3})× the minimum deposition velocity(m s^{-1}) × interval time(s).

A minimum deposition velocity of $1.0 \times 10^{-4} \text{ ms}^{-1}$ was obtained from Yasunari et al. (2010). Ignoring the seasonal variation of the viscosity coefficient of air, we assumed that the minimum deposition velocities in the monsoon and nonmonsoon seasons were the same, thus we could determine a lower limit of TC deposition. Calculated TC deposition flux was over 16 mgm⁻² (18.75 µgm⁻²day⁻¹) during the whole sampling period, and the mean daily deposition flux of BC accounted for 5.8 % of TC.

Snow and ice are sensitive to BC on the surface layer, especially for fresh snow. BC on the surface layer could significantly decrease the albedo of fresh snow, and BC concentration of 15 ngg^{-1} could lead to a 1 % reduction of albedo (Warren and Wiscombe 1980). Therefore, we estimated the lower limit of BC deposition flux and the surface concentration of Zhadang Glacier in different seasons. Zhadang Glacier (30.47° N, 90.5° E; 5,800 m a.s.l.) in Nyainqêntanglha Mountains is approximately 20 km southwest to the NCOS (Fig. 1). BC deposition flux was higher in the nonmonsoon season than that in the monsoon season (Fig. 10), which was primarily related to the seasonal variation of emission sources and regional atmospheric conditions.

Due to the minimum deposition velocity used in the calculation, estimated BC deposition flux would be underestimating the real deposition flux, nevertheless, it was helpful in estimating BC concentrations in the surface snow layer. Here, we only considered dry deposition in the nonmonsoon and monsoon seasons due to the lack of a wet scavenging ratio. Wet deposition should be the main sink in the monsoon seasons due to higher precipitation. BC in the snow layer on the glacial surface by dry deposition was calculated as follows (Yasunari et al. 2010):

$$BC_{snow} = BC_{deposition} \times area / (\rho_{snow} \times area \times thickness_{snow})$$

where thickness_{snow} was set to 2 cm and ρ_{snow} of settled snow was assumed to be 250 kgm⁻³ (Cuffey and Paterson 2010).

Based on the method previously described, the mean daily BC concentration deposited on the glacial surface snow layer from August to October reached 0.69, 0.59, and 1.59 ngg^{-1} in 2006, 2007, and 2009, respectively. High BC concentration in October 2009 increased the mean daily snowpack BC (~1.59 ngg^{-1}) to 3.84 ngg^{-1} , which shows that the episode significantly changed BC concentrations in the snowpack on a seasonal scale which would furthermore affect the surface albedo (Fig. 11a–c). Deposited BC concentration in winter (December–

Fig. 10 BC deposition flux in the monsoon and nonmonsoon seasons, with the *red solid line* for the mean level in the monsoon season, the *blue solid line* for the mean level in the nonmonsoon season, and *red* and *blue dashed lines* for the mean level by ignoring the BC peaks occurring in mid-March and mid-October 2009. M representing for monsoon season and N-M representing for nonmonsoon season

February) has been increasing gradually from 0.65 ngg^{-1} in 2007 to 0.90 ngg^{-1} in 2009 (Fig. 11d–f), which indicates that the atmospheric environment in central TP became worse and the influences from human activities have strengthened.

Conclusions

Atmospheric carbonaceous aerosols were sampled discontinuously from July 2006 to December 2009 at NCOS in central TP. The mean daily concentration of carbonaceous

Fig. 11 BC concentration in the glacial surface snow layer during the same period in different years, each step started and ended at the beginning and ending time

aerosols increased from July 2006 to March 2009, and a high OC/BC ratio indicated that OC was the main component of carbonaceous aerosols. Seven-day air masses backward trajectories calculated by the HYSPLIT model suggested that atmospheric pollutants emitted from Northern India and South Asia could rapidly penetrate into central TP via southwest winds. Carbonaceous aerosols could contribute significantly to the AOD in central TP. The distribution of AOD all over the TP was likely to be associated with the high AOD at the southern slope of the Himalayas. Higher BC deposition flux in the nonmonsoon season than that in the monsoon season primarily resulted from seasonal variations of emission sources and regional atmospheric conditions. Increased BC concentration in wintertime snowpack from 2007 to 2009 indicated that atmospheric environment in central TP became more contaminated and influences from human activities have strengthened. Episodes occurring at pristine and remote sites could significantly change BC concentration in the snowpack on a seasonal scale, which would furthermore affect the surface albedo. Although the effects of carbonaceous aerosols on global/regional climate and environment are very complicated, the analysis of in situ measurements helps us better understand these effects.

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