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Key Points:

- Laser-induced-incandescence measurements revealed no seasonal variability in black carbon (BC) mass size distributions at Jungfraujoch
- BC was heavily coated in summer but largely uncoated in winter, a seasonality that has not been reported before
- These measurements have important implications for the aging timescales and lifetime of BC in the free troposphere

Supporting Information:

- Supporting Information S1
- Data Set S1

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Black Carbon Aerosols in the Lower Free Troposphere are Heavily Coated in Summer but Largely Uncoated in Winter at Jungfraujoch in the Swiss Alps

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Abstract Black carbon (BC) particles are responsible for substantial radiative heating of the atmosphere. However, the climate-relevant properties of BC particles in the free troposphere (FT) are poorly constrained over longer time scales. Here, we report on in situ, intensive field campaigns deploying a single-particle soot photometer during summer and winter seasons at the Jungfraujoch (3,580 m a.s.l., Switzerland), a site often located in the lower FT. The refractory BC (rBC) mass size distribution was very stable across the two seasons. BC was generally internally mixed (i.e., heavily coated with other aerosol material) in summer. However, against general expectations, BC was predominantly externally mixed in winter, with a high occurrence of negligibly-to-thinly coated BC. This strong seasonality in lower-FT BC mixing state has not been previously observed and would substantially influence the lifetime, loading, and climate effects of free-tropospheric BC. Future measurements should investigate the generality of these results.

Plain Language Summary Black carbon (BC) aerosols (i.e., soot particles) play an important role in global climate change by causing atmospheric warming. To understand this role, we need to know what types of BC particles are found in the atmosphere; how big are they and how are they mixed with other aerosols? This type of information is lacking for BC particles found in the free troposphere (the atmospheric layer immediately above the mixed boundary layer of air at the Earth's surface). We performed measurements during winter and summer seasons at Jungfraujoch in the Swiss Alps (3,580 m a.s.l.) in order to fill this gap in knowledge. We discovered a large and unexpected seasonal contrast; in summer at this site, BC aerosols are heavily coated with other aerosol material, whereas in winter the BC is largely uncoated. This seasonal contrast has important implications for our understanding of how long BC particles stay in the atmosphere (coated particles are removed faster), and our estimates of BC climate impacts (coated particles absorb more sunlight).

1. Introduction

Black carbon (BC) is an important component of atmospheric particulate matter. It is a short-lived air pollutant that is responsible for more direct radiative forcing (i.e., forcing due to aerosol-radiation interactions) than any of the greenhouse gases except carbon dioxide and perhaps methane (Myhre et al., 2013). Consequently, the quantification of BC climate forcing has recently gained much attention (e.g., Booth & Bellouin, 2015; Samset & Myhre, 2015). BC forms during the incomplete combustion of fossil and biogenic fuels and mainly originates from anthropogenic sources (Klimont et al., 2017). BC particles acquire so-called “coatings” during their atmospheric life cycle due to the condensation of and/or coagulation with organic and inorganic material. This aging process modifies the absorption of solar radiation by BC (Fuller et al., 1999) and increases the capacity of BC particles to form cloud droplets in supersaturated conditions (Dalirian et al., 2018). The estimated radiative forcing due to BC aerosol-radiation interactions amounts to $+0.6 \text{ W m}^{-2}$ over the industrial era (Myhre et al., 2013). However, the uncertainty in this estimate remains very high (90% uncertainty bounds: $+0.23$ to $+1.02 \text{ W m}^{-2}$).

In order to reduce the large uncertainty in the radiative forcing estimates of BC, it is important to accurately simulate its atmospheric life cycle (Boucher et al., 2016; Hodnebrog et al., 2014). Recent modeling

(Matsui, 2016) and experimental studies (Motos, Schmale, Corbin, Zanatta, et al., 2019; Ohata et al., 2016) have confirmed that coated BC particles have shorter lifetimes than uncoated BC particles, as they are removed more efficiently by nucleation scavenging (cloud droplet formation). Nucleation scavenging efficiency is also a function of particle size, which means BC size distribution also modulates the wet removal and thereby lifetime of BC (Ching et al., 2018; Matsui et al., 2018). This size dependence was recently confirmed experimentally with in situ observations in clouds at the high-altitude research station Jungfraujoch (JFJ) in Switzerland (Motos, Schmale, Corbin, Modini, et al., 2019).

Although the concentration of BC particles is generally lower in the free troposphere (FT) than in the planetary boundary layer (PBL; Lund et al., 2018), free-tropospheric BC particles still contribute substantially to the total direct radiative forcing of BC, as well as the large uncertainties in the estimates of this quantity (Samset et al., 2013). BC particles reach the FT by direct emission at altitude (e.g., from aviation sources) or by transport from sources in the PBL (e.g., biomass burning; Petetin et al., 2018). Model calculations suggest that direct aviation emissions contribute only a minor amount of BC mass to the upper FT, even along busy flight paths (Hendricks et al., 2004). BC transport from the PBL can be accomplished by thermally induced convection, orographic lifting, synoptic-scale winds, and other vertical air movements. The limited available airborne measurements indicate that the fraction of internally mixed BC (i.e., coated BC) increases with altitude through the FT (Hara et al., 2003; Pratt & Prather, 2010; Schwarz et al., 2008), which is consistent with the notion that BC from emission sources in the PBL becomes progressively more aged and coated as it is transported through the atmosphere (McMeeking et al., 2011; Moteki et al., 2007).

Airborne measurements are well suited for characterizing specific pollution plumes (Dahlkötter et al., 2014; Ditas et al., 2018) and obtaining snapshots in time of vertical profiles of BC mass concentrations (Lund et al., 2018; Matsui et al., 2011; Oshima et al., 2012; Schwarz et al., 2013). However, aircraft platforms cannot be used to perform continuous, longer-term, background FT measurements. Additionally, the low concentrations of BC found in the FT make it difficult to obtain statistically robust measurements of BC size distributions and mixing state from fast-moving aircraft. Consequently, the observational picture of the microphysical properties of BC in the background FT is both incomplete and uncertain, particularly over longer (e.g., seasonal) timescales.

The high elevation of the JFJ research station makes it an ideal site for studying aerosol properties in the background lower FT over longer timescales. At 3,580 m a.s.l. the station is often situated completely in the lower FT. At other times it receives variable injections of air masses originating from the PBL depending on the season and the regional weather conditions (Collaud Coen et al., 2011; Herrmann et al., 2015). Numerous BC-related studies have already taken place at the JFJ focusing on, for example, optical properties (Lavanchy et al., 1999), nucleation scavenging (Cozic et al., 2007), and ice nucleation properties (Eriksen Hammer et al., 2018; Kupiszewski et al., 2016). Liu et al. (2010) reported the first single-particle characterization of BC at the JFJ with a single particle soot photometer (SP2). However, the early version of the SP2 that was used in that study measured only a limited size range of BC, and it was not equipped with a detector for quantitative mixing state measurements.

In the present study, we used single-particle measurements from an updated SP2 instrument to characterize and compare the mass concentrations, mass size distributions, and mixing states of BC during winter and summer seasons at the JFJ. Unexpectedly, we observed that the free-tropospheric BC measured in winter was largely uncoated (i.e., externally mixed), in contrast to the heavily coated and aged BC that was observed in the summer FT.

2. Experimental

2.1. Measurement Site and CLACE Campaigns

Stationary in situ measurements were conducted at the high-alpine research station Jungfraujoch (3,580 m a.s.l.) in the Swiss Alps. The JFJ is located on a mountain pass between two high alpine peaks: the Jungfrau (4,158 m a.s.l.) and the Mönch (4,107 m a.s.l.). Continuous aerosol measurements have been conducted for more than 20 years at the JFJ through an aerosol monitoring program that became part of the Global Atmospheric Watch program of the World Meteorological Organization in 1995 (e.g., Bukowiecki et al., 2016; Poltera et al., 2017; Zieger et al., 2012). Results from two CLOUD and Aerosol

Figure 1. Diurnal cycles of BC core mass concentrations for $D_{rBC} > \approx 50$ nm during CLACE2014 (winter) and CLACE2016 (summer). In this figure and in Figure S1, the “FT” and “PBL” classifications are different from the other figures: Whole days (rather than shorter periods) are assigned to one of these two classes as described in section 2.2.3. Total sampling time reflects the total number of hours for which the SP2 furnished valid data behind the total air inlet for a given condition.

Characterization Experiment (CLACE) campaigns are presented in this paper: the CLACE2014 campaign from 17 January to 3 March 2014 and the CLACE2016 campaign from 13 June to 5 August 2016.

2.2. Instrumentation and Data Analysis Approaches

2.2.1. The Total Inlet

All particle instruments were connected to a “total” inlet, which is utilized for the long-term observations and samples air 2 m above the laboratory roof. This inlet collects all aerosol particles, that is, interstitial (non-activated) and droplet residual particles during cloud conditions (Weingartner et al., 1999). It is heated to $\approx 20^\circ\text{C}$ (Nessler et al., 2003) in order to keep the relative humidity below 20% and has been shown to sample hydrometeors with a diameter up to $40\text{ }\mu\text{m}$ at wind speeds up to 20 m s^{-1} .

2.2.2. BC Mass Concentration, Mass Size Distribution and Mixing State

The single-particle soot photometer (SP2 upgraded to eight-channel Revision C version, Droplet Measurement Technologies, Longmont, CO, USA) was utilized for BC characterization. SP2 data can be used to derive refractory BC (rBC) mass, rBC mass equivalent diameter (D_{rBC} ; conversion of rBC mass to D_{rBC} using BC material density of $1,800\text{ kg m}^{-3}$), and BC mixing state (thickness of non-BC coating under a concentric spheres core-shell approximation) on a single-particle level (Gao et al., 2007; Laborde et al., 2012, 2013; Moteki & Kondo, 2007; Taylor et al., 2015). This instrument is briefly described in Text S1. The SP2 scattering detectors were calibrated three to four times per campaign. The incandescence detectors, which are more stable than the scattering detectors, were calibrated once per campaign using the recommended fullerene soot sample (Baumgardner et al., 2012; Laborde et al., 2012). The obtained incandescence calibration curves agreed well with previous calibration curves for the instrument. Although the duration of CLACE2016 was longer than that of CLACE2014, instrument malfunction meant that a greater amount of valid SP2 data was collected during CLACE2014 than CLACE2016.

2.2.3. Particle Number Size Distribution and Criteria for FT Conditions

Particle number size distribution is continuously measured at JFJ using a custom-built scanning mobility particle sizer (SMPS; Wiedensohler et al., 2012). The SMPS was validated every 2 weeks during the campaigns using polystyrene latex size standards (Thermo Fischer Scientific, Waltham, MA, USA).

Herrmann et al. (2015) analyzed 6 years of SMPS data at the JFJ to characterize the particle number size distribution in different types of air masses, specifically investigating in-cloud/out-of-cloud and FT/PBL

conditions. These authors found that the number concentration of particles larger than 90 nm in diameter (N_{90}) is strongly influenced by PBL injections. Therefore, N_{90} is a good parameter for distinguishing between FT conditions and PBL-influenced air masses. For CLACE2014 winter data, we defined FT conditions if N_{90} (6 min resolution) was below 1.5 times the winter-derived baseline of 30 cm^{-3} observed by Herrmann et al. (2015). All data that did not match this criterion were classified as “PBL-influenced.” The same was done for CLACE2016 summer data using the summer baseline (55 cm^{-3}) to define the threshold. When classifying daily profiles, which is done for Figures 1 and S1, the whole day was classified as “PBL-influenced” if the average particle number concentrations exceeded the aforementioned seasonal threshold between 15:00 and 22:00 local time (LT), which is the time period when PBL influence is most apparent, as discussed below (section 3.2). If this criterion was not achieved the whole day was classified as “FT.” Note that days on which the threshold N_{90} level was only exceeded during short peaks of local pollution were excluded from the daily profile analyses. All data were also classified according to “northwest” (NE) and “southeast” (SW) local wind conditions if the wind direction (10 min resolution) was measured in the range $280\text{--}350^\circ$ and $100\text{--}170^\circ$, respectively. Measurements at the JFJ are typically classified into these two wind directions since the station is located on a mountainous ridge that is oriented in a direction from southwest to northeast (e.g., Bukowiecki et al., 2016).

3. Results

3.1. FT Versus PBL-Influenced Conditions

During CLACE2014 in the winter season, FT and PBL-influenced conditions (criteria described in section 2.2.3) occurred roughly 70% and 30% of the time, respectively (Table S1). The situation was opposite in summer (CLACE2016) with 69% PBL-influenced and 31% FT conditions, as expected due to enhanced thermal convection in the summer season, which typically affects the site in the afternoon (Collaud Coen et al., 2011). The prevalence of FT conditions during CLACE2014 and CLACE2016 was similar though slightly more frequent than reported by Herrmann et al. (2015) for the respective seasons based on 6 years of gas measurements (^{222}Rn and NO_y/CO). Southeasterly and northwesterly wind conditions occurred with almost equal frequency in winter (Table S1), while higher frequencies of northwesterly (71%) than southeasterly winds were observed in summer.

3.2. Mass Concentrations and Mass Size Distributions of BC

BC mass concentrations were calculated based on the counting of incandescence signals by the SP2 and application of the empirical incandescence calibration (Text S1). Figure 1 shows diurnal cycles of BC mass concentration during winter and summer. The diurnal profiles of BC number concentration look very similar and are shown in Figure S1. These two figures indicate that PBL-influenced conditions are, as expected, associated with higher mass concentrations than FT conditions because of pollution transport. In addition, BC concentrations are higher during southeasterly than northwesterly wind conditions, as previously reported (Liu et al., 2010; Kammermann et al., 2010).

Figure 1 also provides information about the seasonal variations of BC mass concentration and the cause of its variations. Between around 03:00 and 08:00 LT, the median BC mass concentrations are similar in winter compared to summer. However, between approximately 15:00 and 22:00 LT, PBL influence is most apparent, and median summer concentrations are a factor of 4 higher than median winter concentrations. This can be explained by thermal convection, which carries particles up to the altitude of the JFJ mainly during the afternoon hours, as shown previously (Baltensperger et al., 1997; Lugauer et al., 1998). This characteristic profile is observed most clearly when the wind comes from the northwest. Lugauer et al. (2000) showed that, since the JFJ is located at the northwestern edge of the Alps, the aerosol concentration drops rapidly in the evening after the vertical transport stops for winds from the NW. In contrast, for winds from the SE where further mountain peaks lie, vertical transport may have taken place at substantially larger distances from the JFJ, resulting in elevated aerosol concentrations during extended periods of the night. This results in an increase of the overall aerosol concentrations during SE wind conditions and a flatter diurnal profile, consistent with the SE profiles shown in Figure 1. Because of the occurrence of overcast conditions blocking convection, BC mass concentrations are very variable in summer, as shown by the very large interquartile range (IQR). Winter BC mass concentration profiles at the JFJ are very different to summer ones: They indicate very stable concentrations because thermal convection is weak. Most winter days were thus classified with “FT

Figure 2. Averaged coating thickness histograms (normalized by the area) during the CLACE2014 (winter) and CLACE 2016 (summer) campaigns, (a) for two different ranges of D_{rBC} and (b) filtered by wind direction and PBL/FT criteria.

conditions.” Some winter days were classified as “PBL-influenced,” but the afternoon concentrations were only a factor of ~ 2 higher than the morning concentrations, indicating less variation than what was observed for summer “PBL” days.

The counting and sizing of individual BC cores by the SP2, based on the analysis of incandescence signals, permit the calculation of BC mass size distributions as a function of r_{BC} core mass equivalent diameter (D_{rBC}). Average mass size distributions over the whole CLACE2014 and CLACE2016 campaigns are shown as solid black lines in Figures S2 (absolute concentrations) and S3 (normalized to equal area below the curve). The average distributions for each season peak at very similar D_{rBC} , between roughly 130 and 150 nm, and have similar shapes, indicating that there is very little seasonality in the BC mass size distribution at JFJ. Differences in the mode of the averaged winter and summer BC mass size distributions during FT conditions are not greater than experimental uncertainty, which is affected by limited counting statistics in the summer data set. The variations in the shape of the BC mass size distributions due to the wind direction and the air mass type (PBL/FT) are also relatively small, as shown in Figure S3. Figure S2 shows that average BC mass concentrations are a factor of 2 higher in summer than in winter; this applies to all types of air mass criteria, except for FT conditions. Liu et al. (2010) also reported SP2-derived BC mass size distributions at the JFJ in winter, and our results are in good agreement with their findings for D_{rBC} above 190 nm (Figure S2), while only the newer SP2 used in this study was able to fully resolve the leading tail and modal diameter of the r_{BC} mass size distribution (see Text S3 for further discussion).

3.3. Strong Seasonality of BC Mixing State

Quantitative information on BC mixing state was obtained using the LEO-fit technique (see Text S1), which allows for the derivation of coating thickness histograms. To provide confidence in the LEO-fit results, we first compared them against results extracted from the delay time method (Text S1), a qualitative but more robust method providing the number fractions of BC cores classified as thinly-to-moderately coated or thickly coated. Figure S4 compares the products of these two methods for 8 days, chosen to cover the range of mixing state variation observed within each season. There is excellent qualitative agreement between the results from the delay time and LEO-fit techniques: For instance during winter, large fractions of thin to moderate coatings are associated to coating thickness histograms that peak around 0 nm and contain very few occurrences of coatings larger than 40 nm, indicating externally mixed BC is dominant. By contrast,

the BC population was largely internally mixed during summer, with large fractions of thickly coated particles that are associated to coating thickness histograms that peak at values greater than 20 nm and contain many occurrences of coatings in the range from 40 to 120 nm.

The quantitative coating thickness analysis based on the SP2 LEO-fit technique was extended to the whole CLACE2014 and CLACE2016 campaigns in order to obtain results that are as representative as possible for the winter and summer seasons. The coating thickness histograms are depicted for two different size ranges in Figure 2a and are filtered by NW/SE and PBL/FT criteria in Figure 2b. These results confirm the substantial difference in BC mixing state between winter and summer conditions observed for the eight example days shown in Figure S4. They indicate a strong degree of external mixing in winter with histograms peaking around 0 nm and very few particles with a coating larger than 40 nm, contrasting with the prevalence of coatings between 20 and 70 nm in summer. This finding is consistent across the two ranges of BC core mass equivalent diameters that were examined ($D_{\text{rBC}} = 200 \pm 20$ nm and $D_{\text{rBC}} = 250 \pm 30$ nm; Figure 2a; note that at smaller and larger D_{rBC} coating thickness histograms are truncated due to SP2 detection limits and are therefore not suitable for quantitative comparisons). Similarly, the seasonal contrast is also consistent across the NW/SE wind and PBL/FT air mass categories that were examined (Figure 2b).

4. Possible Reasons for the Strong Seasonality in BC Mixing State

Given its remote alpine location and apparent distance from fresh BC source regions, it is expected that the BC particles reaching the JFJ will be aged and relatively thickly coated, as observed during the summer season. The observation of stable concentrations of largely uncoated BC at the JFJ during winter was unexpected and can be explained by several possible hypotheses. First, it is important to note that the average coating thickness histograms displayed in Figure 2 were not influenced by specific event(s) of very high BC concentrations. Additionally, short, sporadic periods of particulate emissions from construction work at the JFJ station were strictly reported and excluded from the analysis. Therefore, we consider that the sampling durations of CLACE2014 (27.9 days) and CLACE2016 (26.8 days) were sufficiently long and unaffected by specific and/or local events to be seasonally representative. Second, possible SP2 measurement and analysis artifacts, as well as possible sampling biases between the two seasons, were systematically considered and ruled out as explanations for the observations in Figure 2, as discussed in Text S2. In addition, the comprehensive wintertime mixing state measurements presented in this study were compared with short-term measurements from previous winter seasons at JFJ, as discussed in detail in Text S3 (Liu et al., 2010; Kupiszewski et al., 2016). Most importantly, the results presented in this study are consistent with the in-cloud, wintertime measurements of Kupiszewski et al. (2016), which also indicated a prevalence of externally mixed BC (Figure S5).

We pose and evaluate the following hypotheses to explain our winter BC mixing state measurements: (1) The BC measured during winter at JFJ originated primarily from sources above the PBL such as ski resorts, restaurants, and hotels; (2) the BC originated mainly in the PBL but remained uncoated during transport to the site, which potentially occurred over long ranges; or (3) heavily coated BC was preferentially removed from air masses transported to the JFJ during winter (by, e.g., nucleation scavenging), leaving behind a high fraction of uncoated BC particles in the FT air. The three hypotheses are not mutually exclusive, and a combination of these processes may have occurred.

Regarding Hypothesis 1, the diurnal cycles of BC concentrations (Figure 1) during PBL-influenced conditions in winter indicated that these periods were associated with large-scale synoptic winds rather than strong vertical air movements associated with thermally induced convection. In winter in the European Alps there are several potential sources of fresh BC emissions located above the PBL at altitudes up to 2,500–3,500 m a.s.l, including engines, generators, and wood stoves in ski resorts, restaurants, and/or hotels. We performed a rough calculation with emission rate estimates from snow groomers in Switzerland only, which indicates that these machines alone could potentially contribute three quarters of the BC mass in the FT above the Swiss Alps during winter (results and details of the calculation both described in Table S2). Therefore, we consider it plausible that fresh emission sources of BC above the PBL could contribute substantially to the concentrations of externally mixed BC particles observed at the JFJ during winter. This potential phenomenon is of regional importance, since high-altitude BC emission sources extend over

the entire European Alps and BC emitted directly into the FT can be expected to have longer average life-times than BC emitted in the PBL, where removal processes are generally more active.

It is also possible that during winter, BC can be transported to the JFJ from potentially distant sources within the PBL without acquiring substantial coatings, which leads to the formulation of Hypothesis 2. This would be possible if the BC aging (i.e., coating acquisition) process was very slow during winter conditions: that is, if BC aging timescales were greater than the time required for BC transport to the JFJ, which could vary from hours and days (continental sources) up to weeks (intercontinental sources) (e.g., Lööv et al., 2008). Several independent studies have suggested that BC aging is dominated by coagulation during the night (which is a slow process with timescales on the order of days to weeks) and condensation during the day (a quicker process with timescales of approximately days), mainly due to the absence of photochemistry during the night, which is required to produce important condensable vapors such as H_2SO_4 via OH oxidation (Doran et al., 2008; Fierce et al., 2015; Riemer et al., 2004, 2010). Liu et al. (2011) suggested that in analogy to this diurnal contrast, reduced photochemistry would also lead to a seasonal contrast in BC aging processes, with the slower coagulation process becoming dominant during winter. These authors found that this seasonal contrast in aging timescale had to be taken into account in order to capture the observed seasonal variations in Arctic BC concentrations in a global climate model. Similar results were obtained with a separate model setup by Lund and Berntsen (2012). However, both of these studies only modeled condensational growth due to H_2SO_4 . Nitrate (Eriksson et al., 2017; Riemer et al., 2004) and secondary organics (He et al., 2016) are additional sources of condensable material that can coat BC particles and thereby speed up the aging process. The seasonal variations in the concentrations of these condensable species at the JFJ can vary considerably from what is expected at lower altitudes in the PBL (e.g., Fröhlich et al., 2015), which complicates assessment of the role that each species might play in modulating BC aging timescales. To unravel these roles and further assess Hypothesis 2, additional measurements of the chemical composition of BC coatings at JFJ would be useful.

To further examine Hypothesis 2, we compiled a subset of previously published BC coating thickness measurements in Figure S6, in order to assess the prevalence of externally mixed BC in the winter PBL. For comparison with our JFJ measurements, we focused this compilation on measurements taken during “background” conditions (i.e., excluding studies focused on biomass burning plumes or other specific pollution events). This summary indicates that high fractions of bare, uncoated BC are indeed common in the background PBL, even if very thickly coated BC particles are also frequently present. This supports Hypothesis 2 as a possible explanation for the large fraction of externally mixed particles observed at the JFJ during winter.

Finally, the high proportion of bare BC remaining in the FT air measured at the JFJ might be the result of the preferential wet removal of heavily coated BC during transport to the site—Hypothesis 3. This explanation is based on the fact that nucleation scavenging is more efficient for coated than uncoated BC particles (Motos, Schmale, Corbin, Zanatta, et al., 2019). However, nucleation scavenging is also a size-dependent process (Moteiki et al., 2012, 2019; Ohata et al., 2016). Therefore, if preferential removal of some fraction of the BC aerosol did occur, one would also expect to see a change in the shape of the BC mass size distribution before and after the removal process. Since the size distributions of the BC particles at emission (i.e., before any potential removal process has acted) are unknown, we instead compare the measured winter and summer BC mass size distributions. These distributions were very similar in shape across the two campaigns (Figure S3). Therefore, we consider it unlikely that the preferential removal of heavily coated BC was responsible for the high fraction of externally mixed BC observed during winter. If this were the case, one would expect to see a similar process occurring in the summer, given the similarity in the shapes of the seasonal BC mass size distributions. Instead, high proportions of heavily coated BC particles were observed during summer, which suggests that preferential wet deposition was not an important process.

Dedicated model calculations are required to finally determine the main reason(s) for the seasonality in BC mixing state observed at the JFJ and to investigate if a similar seasonality occurs more generally in the FT. The value of the measurements presented here is that they were not driven by a specific event or process. This means that even large-scale chemical transport or Earth system models could be used to explore this question, even though these models may only be able to resolve monthly averaged quantities. Such modeled quantities would be comparable to our longer-term, averaged observations. To complement these model

calculations, additional measurements of the chemical composition of BC coatings should be performed to better understand the observed seasonality in coating amount.

5. Conclusions

The seasonal mass concentrations, mass size distributions, and mixing states of refractory black carbon (rBC) were measured and investigated at the high-alpine site Jungfraujoch (JFJ) in winter 2013/2014 and summer 2016 (CLACE campaigns). Summer early morning BC mass concentrations were similar to all-day winter concentrations, indicating the prevalence of FT conditions during these time periods, as shown previously (Herrmann et al., 2015). During summer afternoons and evenings, thermally driven convection caused frequent mixing of BC from low-altitude sources into the air sampled at the JFJ, which resulted in elevated BC concentrations. The mean mass equivalent diameters of BC cores were similar during both seasons, and in the range from 130–150 nm.

During summer, internally mixed BC was dominant at the JFJ (i.e., heavily coated). However, during winter, the majority of BC particles were found to be externally mixed (i.e., uncoated or only very thinly coated). This is the first direct observation of a strong seasonality in the mixing state of free-tropospheric BC. We suggest this seasonality results from slow BC aging times during winter, and/or the presence of BC emission sources above the wintertime PBL in the European Alps, rather than the preferential removal of heavily coated BC particles by wet deposition during wintertime transport to the JFJ. Model calculations are required to confirm this. In addition, model calculations and measurements at additional sites should be performed to explore the generality of our seasonal observations and to assess the resulting impacts on the atmospheric lifetime and climate effects of BC.

Data Availability Statements

The data archive for this manuscript is available on Zenodo (<https://doi.org/10.5281/zenodo.3950037>).

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