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Evaluation of a Perpendicular Inlet for Airborne Sampling of Interstitial Submicron Black-Carbon Aerosol

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The majority of airborne aerosol measurements employ forward-facing inlets with near-isokinetic sampling; these inlets have known artifacts when sampling in clouds such that data taken in cloud must typically be discarded. Here we report first results from a perpendicular inlet for sampling interstitial submicron black-carbon (BC) containing aerosol. The inlet, consisting of a flat plate to stabilize flow prior to perpendicular sampling, was evaluated using a single particle soot photometer (SP2) aboard the NASA WB-57F aircraft during the Midlatitude Airborne Cirrus Properties Experiment (MACPEX) of 2011. The new inlet rejects large particles and is free of aerosol artifacts when sampling in ice clouds while allowing sampling of submicron BC-containing aerosol with the same unit efficiency as a validated isokinetic inlet, thus allowing for airborne sampling of interstitial BC aerosol.

INTRODUCTION

Atmospheric aerosols have wide-ranging impacts in the global environment. They affect the radiation budget of the earth directly through absorption and scattering of radiation, and indirectly through their role in cloud formation (Charlson et al. 1992; Haywood and Boucher 2000; Ramanathan et al.

2001); also, aerosols also have important ramifications for human health (Lighty et al. 2000). Many studies in recent decades have made use of airborne observations of aerosol and much work has been done to develop and characterize inlets for the sampling of aerosols of a wide range of sizes from aircraft (Hangel and Willeke 1990; Huebert et al. 1990; Blomquist et al. 2001; Hermann et al. 2001; Hegg et al. 2005; Bahreini et al. 2008; von der Weiden et al. 2009). Airborne sampling of aerosol is complicated by typically high velocities of aircraft and non-negligible inertia of the particles, which leads to sampling efficiencies that depend on particle mass and size, and sampling pressure (and therefore altitude). A common solution to this problem uses forward-facing inlets with isokinetic sampling followed by passive diffusion, which reduces the aerosol velocity relative to the aircraft without causing significant turbulent aerosol loss, and provides unit sampling efficiency for aerosol diameters over a limited size range typically covering the accumulation mode. These inlets have the problem when sampling in clouds that artifacts are generated when liquid or ice cloud particles collide with inlet surfaces (Weber et al. 1998; Murphy et al. 2004; Schwarz et al. 2006). These artifacts have generally led investigators to remove data taken in clouds and, as a result, few observations of interstitial aerosol from airborne platforms exist in the literature.

A successful interstitial inlet must (1) exclude cloud particles (typically larger than $2\ \mu\text{m}$), (2) be free of artifacts both in and out of clouds, and (3) have known sampling efficiency for the particles of interest. Here we report the use of a perpendicular airborne inlet, previously shown to exclude cloud particles (Popp et al. 2004), to measure small ($<1\text{-}\mu\text{m}$ diameter) particles. We provide evidence that the perpendicular inlet is free of artifacts when sampling BC-containing particles in clouds and evaluate its sampling efficiency for these particles through

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comparison of observed mass distributions and vertical profiles with those obtained by a previously validated (Jonsson et al. 1995; Schwarz et al. 2006), passive near-isokinetic forward-facing inlet using a Single Particle Soot Photometer (SP2, Droplet Measurement Technology, Inc.) aboard the NASA WB-57F aircraft.

METHODS

The Midlatitude Airborne Cirrus Properties Experiment (MACPEX) was an airborne field campaign designed to increase understanding of cirrus cloud properties, processes, and radiative impacts. The present manuscript discusses 12 flights of the NASA WB-57F from Houston, Texas, during March and April of 2011, with most of the in-cloud data collected between 8 and 14-km altitude. All of the clouds encountered with the perpendicular inlet were sampled at temperatures less than 240 K, near the temperature at which homogenous freezing is expected for pure water (235 K). Additionally, the Small Ice Detector 3 instrument (SID3 described below) detected no liquid particles during the periods under discussion. Therefore, the following analysis is representative only of inlet behavior in ice clouds and the results cannot be extended to liquid clouds. Particles containing refractory BC were detected using the NOAA SP2, configured as described by Schwarz et al. (2008a). The term BC is used here to denote carbonaceous material detected by the SP2 that is composed of carbon-spherule aggregates produced during combustion of carbon-based fuels, strongly absorbent of light, and has a vaporization temperature near 4,000 K (Bond et al. 2013).

The instrument, briefly, consists of an intense intra-cavity Nd-YAG laser and a set of detectors to record light that is scattered or emitted by individual particles as they pass through the laser beam. A BC-containing particle absorbs laser light, any associated non-BC material (generically referred to as “coating”) is vaporized, and the BC core is further heated to its vaporization temperature ($\sim 4,200$ K). The hot BC emits thermal radiation, in amounts proportional to its mass, which is imaged onto two photomultiplier tubes (PMTs). In the NOAA SP2 setup, the PMTs have different cathode materials and, hence, respond to different spectral regions of the incandescence. The ratio of the two signals (hereafter denoted the incandescence ratio) can thus be related to the color temperature of the material and is used in post-processing to identify and reject signals arising from incandescence from non-BC populations. The relative gain of the two PMTs is set to give an incandescence ratio of 1 for refractory BC material.

The SP2 as configured here detects individual particle refractory BC masses in the size range of 0.7–130 fg, equivalent to 90–520 nm volume-equivalent diameter (VED) assuming a void-free density of 1.8 g/cm^3 . Note that this diameter range refers only to the BC component, or BC core, of the particle. Ambient BC in the free troposphere is primarily observed as an internal mixture with nonrefractory materials (generically referred to as “coatings”) in which case the actual diameter of the particle can be considerably larger than the volume-equivalent

diameter of the BC component alone. The SP2 can identify the presence of coatings and to estimate coating thickness on a subset of BC sizes as described by Schwarz et al. (2008b) and Gao et al. (2007). Total particle size is determined by scattered 1,064-nm light and is generally found to be in the submicrometer range for the majority of BC-containing particles. The sample flow through the SP2 was 0.24 L/min, volumetric, and there was an additional 2.16 L/min bypass flow for a total flow through the SP2 sampling line of 2.4 L/min. A sample flow of 0.24 L/min increases the particle sampling rate without affecting the focusing of the particle stream (Schwarz et al. 2006).

In addition to the SP2 data, we use meteorological (pressure and temperature) and location (latitude, longitude, and altitude) data provided by the Meteorological Measurement System (MMS) (Scott et al. 1990). Ice and liquid cloud particle data were provided by the SID3 (sizes of 1 to 100 μm) (Cotton et al. 2010), the Video Ice Particle Sampler (VIPS) (sizes of 10 to 200 μm) (McFarquhar and Heymsfield 1996) and the 2D-Stereo probe (2DS) (sizes of 5 to 255 μm) (Lawson et al. 2006). The cloud particle data were used to distinguish between in-cloud and clear-air sampling. All three instruments detect supermicron particles, which are excellent indicators of clouds. In practice, in-cloud sampling periods were defined to be when two of the cloud probes indicated concentrations greater than 10 L^{-1} .

The Forward-Facing Near-Isokinetic Inlet

The forward-facing near-isokinetic inlet, referred to here as the “near-isokinetic inlet” and shown in the top panel of Figure 1, is configured as described by Wilson et al. (1992), Jonsson et al. (1995) and Schwarz et al. (2006). It was mounted on the underside of the aircraft for 4 of the 12 research flights and extended ~ 30 cm from the fuselage. The main inlet body consists of a primary diffuser with an exhaust port, the diameter of which was chosen to achieve isokinetic sampling from the ambient flow. Sample air is heated in the primary diffuser through compressional heating as the relative velocity slows from the ~ 150 m/s speed relative to the WB-57F to ~ 3 m/s within the inlet. Flow sampled through a secondary diffuser, housed within the primary diffuser, is delivered to the SP2. Flow through the primary diffuser is measured using differential pressure ports at the back of the inlet body and that through the secondary diffuser is controlled to a fixed value using a volume flow controller in the SP2 set to maintain isokinetic flow at the tip of the secondary diffuser under typical flight conditions. The flow profile in the inlet was experimentally determined by Wilson et al. (1992). Experimentally determined size- and pressure-dependent transmission efficiencies between the secondary diffuser and the instrument are reported in Schwarz et al. (2006). Briefly, the transmission of all submicron particles is better than 90% at pressures greater than 200 mbarr. At pressures below 200 mbarr, losses are up to 10% for particles less than 700 nm and up to 50% for particles as big as a micron. The NOAA near-isokinetic inlet has successfully been used to measure BC aerosol in cloud-free air on several previous missions aboard the WB-57F (Schwarz et al.

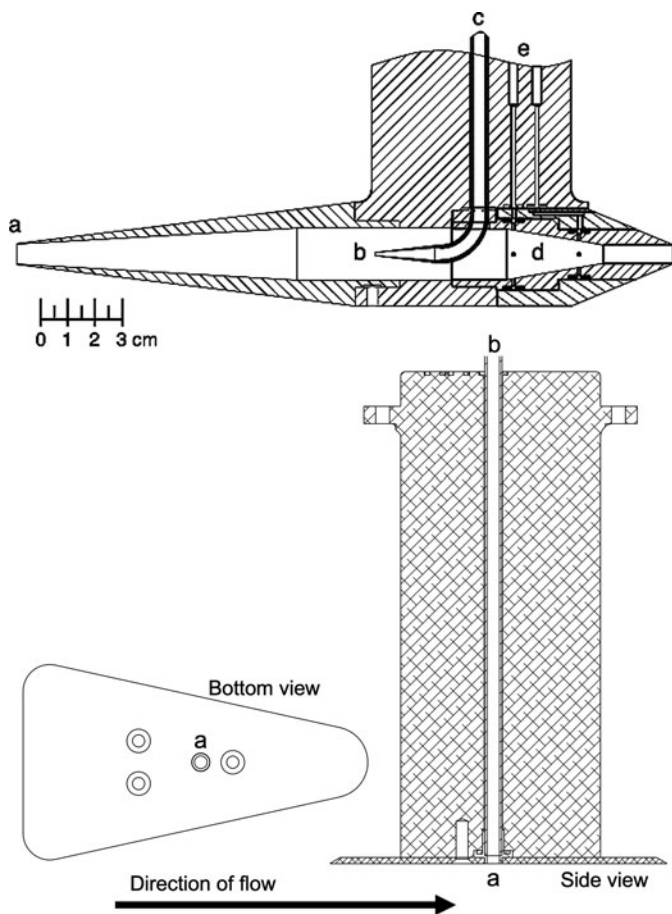


FIG. 1. (Top) Diagram of the near-isokinetic inlet with letters denoting the primary diffuser (a), secondary diffuser (b), flow to the SP2 (c), exhaust port (d), and differential pressure ports (e) taken from Schwarz et al. (2006). (Bottom) Diagram of the perpendicular inlet with letters denoting the inlet port (a) in the flat plate and on the pylon and the inlet line connecting to the SP2 (b). Both inlets are shown with the same scale and are configured with a similar pylon design with the sampling points separated from the fuselage by 30 cm.

2006; Gao et al. 2008; Schwarz et al. 2008a) and on the NOAA P-3D (Schwarz et al. 2008b; Spackman et al. 2008), as well as total submicron aerosol on the NOAA P-3D (Brock et al. 2011).

As stated above, the advantage of an isokinetic inlet is that the sampling efficiency of particles entering the primary diffuser opening is near unity for a wide range of sizes because there is little perturbation of particle trajectories. One disadvantage is that forward-facing inlets have known artifacts when sampling in clouds due to impaction of liquid or ice cloud drops on inlet surfaces (Weber et al. 1998). The impaction can release particles of inlet material such as iron or aluminum, which have been identified during in-cloud sampling by the particle analysis by laser mass spectrometry (PALMS) instrument (Murphy et al. 2004), or material adhered to the inlet surfaces (e.g., BC). As a consequence, data acquired during periods of cloud sampling have unknown contamination by both BC and other particles and have typically been excluded from archived datasets. The

SP2 when previously sampling in clouds has detected high concentrations of incandescent particles, which were identified as non-BC through their anomalous color temperatures and were likely abraded from the inlet surface. In conjunction with this the SP2 often sees anomalously high BC concentrations when sampling in clouds, which do not correlate with concurrent measurements of CO. We attribute these to resuspension of BC that has accumulated on exposed inlet surfaces.

The New Flat-Plate Perpendicular Inlet

The new perpendicular inlet assessed here (shown in the bottom panel of Figure 1) is similar to an inlet originally designed for use with the NOAA Chemical Ionization Mass Spectrometer (CIMS) aboard the NOAA WP-3D and the NASA WB-57F (Neuman et al. 2002; Popp et al. 2004). It was used during 8 of the 12 research flights, in place of the near-isokinetic inlet. Note that only one of the two inlets was used on any given flight and comparisons shown below are, therefore, statistical rather than side-by-side. The comparisons are relevant only to the aspiration into the inlet line opening since inlet-line components between the opening and the SP2 are the same for both inlets, thereby ensuring that internal transmission efficiencies are the same as those reported by Schwarz et al. (2006).

The perpendicular inlet was mounted on the WB-57F at the same location as the near-isokinetic inlet and also extended ~ 30 cm out from the fuselage. The perpendicular inlet is configured around a 4.6-mm inner diameter sample line with its axis oriented perpendicular (90 degrees) to the lower fuselage surface and the ambient flow around the aircraft, and through which air is drawn into the SP2. The flow past the inlet opening is straightened and stabilized with a flat plate mounted parallel (0 degrees) to the lower fuselage surface and the flow on the end of the inlet pylon (Figure 1). The sample speed in the inlet, determined as above by a volumetric flow controller within the SP2, is approximately 3 m/s compared to typical relative speeds of the ambient flow of 150 to 200 m/s with respect to the aircraft.

Popp et al. (2004) reported observations of HNO_3 in contrails from both a forward-facing and a similar perpendicular inlet. They found practically no HNO_3 in the channel sampling from the perpendicular inlet and enhanced HNO_3 in the channel sampling from a forward-facing inlet, which they attributed to HNO_3 condensation on contrail particles, which were then sampled by the forward-facing inlet and excluded from the perpendicular inlet. From these data they inferred that contrail particles larger than approximately $1 \mu\text{m}$ in diameter were inertially stripped from the sample flow in their perpendicular inlet due to the abrupt change in particle velocity and trajectory required in sampling. The lack of HNO_3 signal in the perpendicular inlet channel also indicates that any shattered particle fragments were not sampled.

Although the inner diameter of the sample tube used in the Popp et al. study (4.0 mm) was slightly smaller than that used here (4.6 mm) and the flow rate larger (1.8 standard L/min or approximately a factor of 3.5 higher at 140 mb and 210 K than

the 2.4 volumetric L/min flow used in the present study), simple empirical calculations indicate that cloud particles (which are typically considerably larger than $1\ \mu\text{m}$) should be similarly excluded from the sampled air in our perpendicular inlet. For example, the calculated stopping distance for a $3\text{-}\mu\text{m}$ particle (density of $1\ \text{g/cm}^3$) at a typical air speed ($150\ \text{m/s}$), temperature ($220\ \text{K}$), and pressure altitude ($140\ \text{mb}$ at $14\ \text{km}$) is $\sim 0.75\ \text{cm}$ or almost twice the inner diameter of the sample inlet opening. Hence, it is very likely that such a large particle will not enter the inlet opening or will collide with the inlet wall, representing a loss. The stopping distance for a $10\text{-}\mu\text{m}$ particle is $6.7\ \text{cm}$, which is longer than the distance from the leading edge of the inlet plate to the inlet opening, indicating that such a particle would not even be captured in the boundary layer of the inlet plate. In contrast, the calculated stopping distance for a 100-nm BC particle (density of $1.8\ \text{g/cm}^3$) is $\sim 0.017\ \text{cm}$ or $<5\%$ of the inner diameter of the inlet opening indicating a much higher sampling efficiency for smaller particles.

Determining the exact cut size of the perpendicular inlet is challenging because of the small inlet opening and the large ratio of the free-stream velocity ($150\text{--}200\ \text{m s}^{-1}$) to internal sample flow velocity of between 60 and 80. In addition, there may be boundary layer effects, which are difficult to simulate. The inlet sampling efficiency for such flow conditions has not been addressed in previous laboratory studies or theoretical treatments. A full computational fluid dynamics evaluation of the inlet configuration is beyond the scope of the present effort. In what follows observational evidence is used to infer the sampling efficiency of the perpendicular inlet for the range of BC-containing particle sizes detected by the SP2.

EXPERIMENTAL RESULTS

Artifacts When Sampling in Clouds

Figure 2a shows cloud artifacts from the near-isokinetic inlet when clouds were encountered (indicated by gray shading)

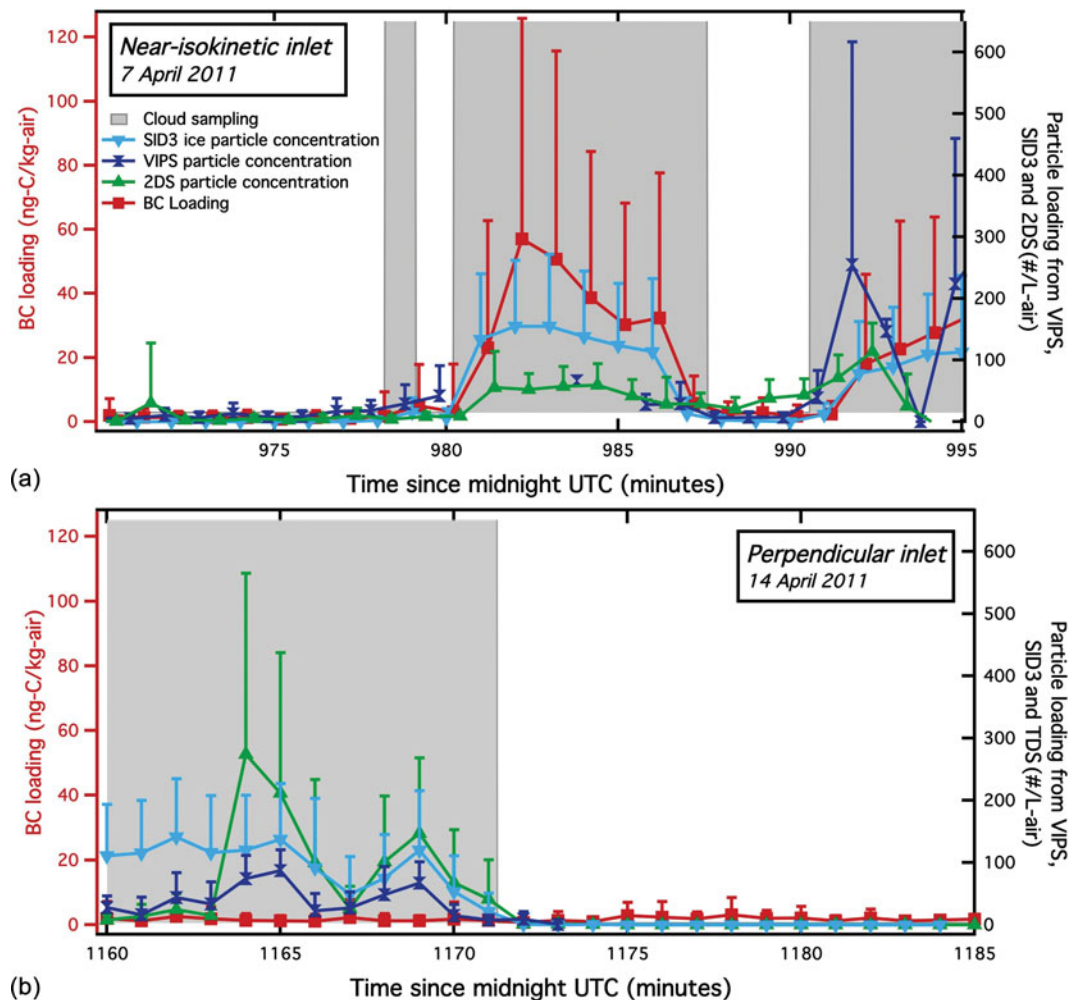


FIG. 2. Sample of in-cloud and out-of-cloud data averaged to 1 min for the near-isokinetic inlet (a) and the perpendicular inlet (b) including VIPS ice probe counts, SID3 ice probe counts, 2DS particle counts (all plotted on right axis), and BC mass loading (left axis). Bars show standard deviation of 1-s data. In-cloud sampling is indicated by gray shading as defined by periods when any two of the cloud probes indicated greater than 10 particles/(L of air). (Color figure available online.)

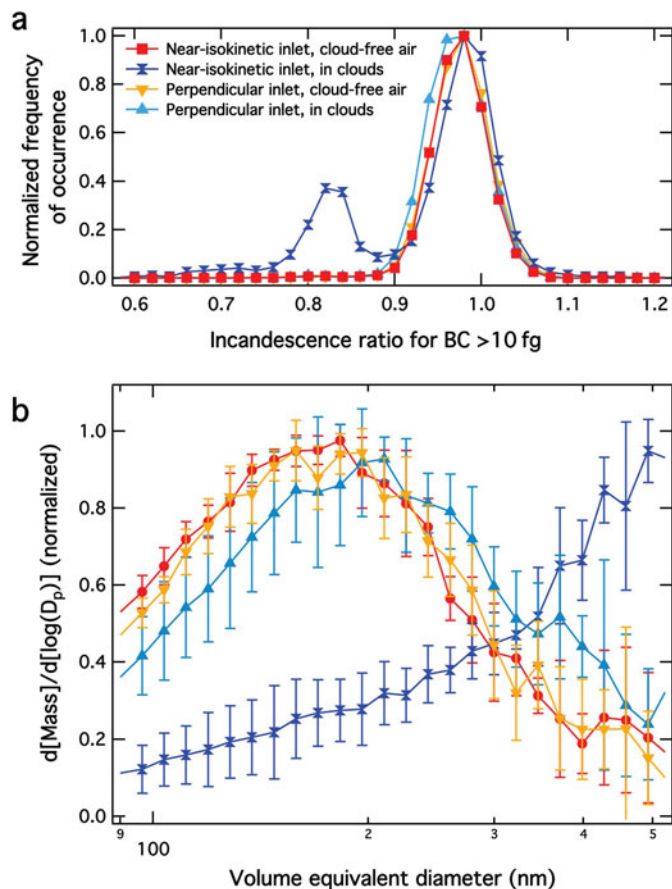


FIG. 3. Normalized histograms of observed incandescence ratios for large (>10 fg) BC-containing particles (a) and normalized mass distributions (b) observed with the near-isokinetic inlet in clouds and out of clouds and with the perpendicular inlet in clouds and out of clouds. Error bars in (b) show the standard deviation of individual distributions calculated for each flight at altitudes above 8 km. Data from all four flights with the near-isokinetic inlet and eight flights with the perpendicular inlet are included. Total particle counts were 1.2×10^5 and 9.5×10^4 for the near isokinetic inlet in and out of clouds, respectively, and 2.6×10^5 and 5.1×10^5 and for the perpendicular inlet in and out of clouds, respectively. Each curve was normalized to have equal peak values in the diameter range shown. (Color figure available online.)

while the bottom panel shows no artifacts from the perpendicular inlet during different cloud encounters. The near-isokinetic inlet data clearly show a response as immediate and anomalously high concentrations of BC are observed when clouds are encountered. The perpendicular inlet does not show this behavior. The justification for identifying this behavior as anomalous is shown in detail in Figure 3. The top panel shows histograms of incandescence ratios of large particles sampled with each inlet both in and out of clouds. Under cloud-free conditions all particles sampled with either inlet lie within a population having an incandescence ratio distributed about 1 (appropriate to BC). In contrast, when sampling with the near-isokinetic inlet in clouds, there is a clear secondary population of particles with an anomalously low incandescence ratio, consistent with detection of metal particles abraded from inlet surfaces. Concurrent

with this, there is also a sudden increase in particles with incandescence ratios indicative of “normal” BC, consistent with resuspension of BC particles deposited to inlet surfaces in the near-isokinetic inlet. This is evidenced by spikes in BC mass loading in clouds (line with squares in Figure 2a), which cannot be entirely accounted for by particles with anomalous incandescence ratios and are uncorrelated with CO. The perpendicular inlet does not produce either of these effects.

Figure 3b shows BC-only mass size distributions sampled with both inlets in and out of clouds at altitudes above 8 km. The cloud-free data for both inlets show a typical BC mass distribution centered near 200-nm VED assuming 1.8 g/cc void-free density. The in-cloud mass distribution when sampling with the near-isokinetic inlet clearly shows anomalously high contributions from large particles and lacks the normal mode entirely. The only known and plausible source of these additional particles is abraded metal and BC particles from inlet surfaces. The in-cloud mass distribution observed with the perpendicular inlet, on the other hand, is log-normal and shows no evidence of a similar-sized artifact response during cloud sampling.

Sampling Efficiency of BC-Containing Particles

The BC-containing particle sampling efficiency of the perpendicular inlet was assessed by comparing observed size distributions and mass loadings with those obtained using the near-isokinetic inlet outside of clouds. From Figure 3, we can clearly see that there are negligible differences between the mass distributions observed with the two inlets under cloud-free conditions and conclude that the perpendicular inlet is not subject to differential losses for BC-containing particles. As noted above, BC cores, which are internally mixed with nonrefractory material, can be substantially larger than the volume equivalent diameter of the BC component alone. This coating information can be used to further evaluate the potential inlet losses or artifact response. As such, we see no evidence for differential losses in comparisons of the two inlets based on (1) similar fractions of particles identified as coated and (2) similar coating thickness calculated for BC cores of 6–8 fg (185–205 nm VED).

Figure 4 shows the composite, mean vertical profiles of BC mass loadings for each inlet. The composite profiles were obtained from individual ascent and descent profiles by first averaging each profile into 1,000-m altitude bins and then averaging all profiles from each inlet. The composite profile for near-isokinetic inlet comprises 8 individual profiles and that for the perpendicular inlet comprises 16. Note that, since only one of the two inlets was used during any given flight, this is a statistical rather than a side-by-side comparison. All data points in the composite profiles agree to within the standard deviation of the mean indicating that the perpendicular inlet is as efficient at sampling accumulation-mode BC-containing aerosol as the near-isokinetic inlet to within the natural variability of the profiles. Ascending and descending profiles were examined to determine potential effects of attack angle and no significant differences were observed. The error-weighted correlation

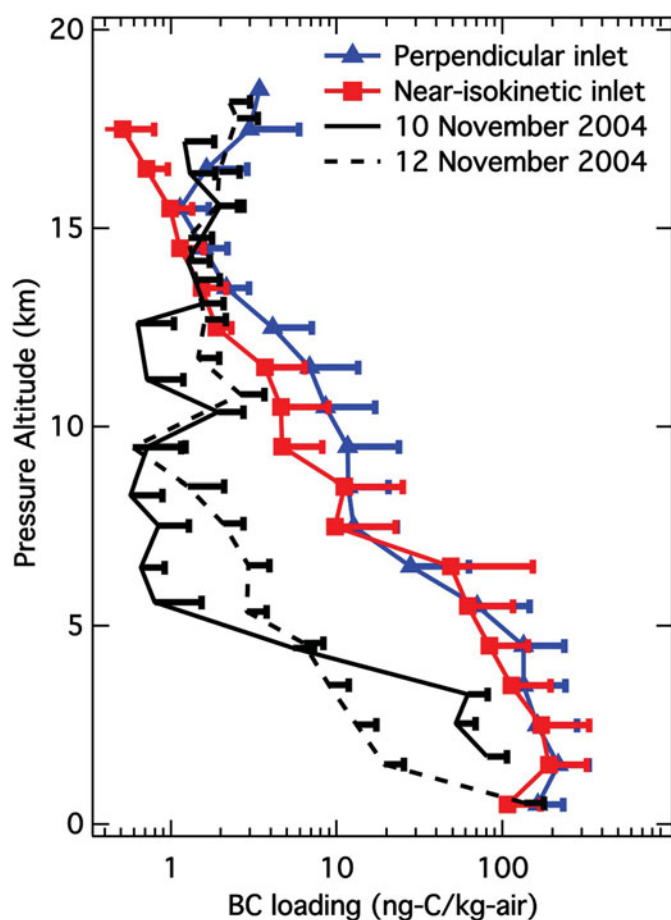


FIG. 4. Composite vertical profiles of BC mass loadings from averages over 1000-m altitude bins observed under clear sky conditions using the near-isokinetic inlet (squares) and the perpendicular inlet (triangles) during the MACPEX mission. Bars show the standard deviation of the individual profile data points at each altitude interval. Seven profiles were obtained using the near-isokinetic inlet and 16 were obtained using the perpendicular inlet. Also shown for comparison (solid and dashed lines) are vertical profiles from two flights in 2004 using the near isokinetic inlet aboard the WB-57F (Schwarz et al. 2006). (Color figure available online.)

between the profile obtained with the perpendicular inlet and that with the near-isokinetic inlet has a slope of 1.36 ± 0.5 at the $1-\sigma$ confidence level (not shown). Shown in Figure 4 for comparison are profiles obtained aboard the WB-57F aircraft from the same SP2 instrument and the same airport during two flights in 2004 with the near-isokinetic inlet. The 2011 loadings are higher than the 2004 loadings, likely because of biomass burning that was occurring in Texas at the time of the MACPEX mission (NOAA National Climate Data Center 2011). In 2004, differences between two BC profiles obtained with the near-isokinetic inlet were attributable to natural, flight-to-flight variability (Schwarz et al. 2006). Since these differences are greater than those between the average profiles obtained with different inlets in 2011, we consider the 2011 agreement to be excellent and take this, along with the agreement between the mass distributions, as ex-

perimental evidence of acceptable sampling efficiency for small BC-containing aerosol particles by the perpendicular inlet.

SUMMARY AND CONCLUSIONS

A perpendicular inlet has been successfully flown aboard the NASA WB-57F for the measurement of small (<580 -nm VED) BC-containing particles both in and out of ice clouds. Previous work and empirical calculations of stopping distance indicate that this inlet excludes cloud particles, which are typically much larger than $1 \mu\text{m}$. In addition, the perpendicular inlet does not generate a discernible artifact response in ice clouds, which is more typical of near-isokinetic diffuser inlets. No significant difference was observed in either BC mass loadings or mass distributions when compared to a previously validated, near-isokinetic, diffuser inlet in cloud-free air. The correlation between loadings observed with the two inlets was 1.36 ± 0.5 indicating that the two inlets have very similar sampling efficiencies over the size range of particles detected by the SP2. Thus, this simple inlet configuration will allow for improved sampling of interstitial BC-containing aerosol in future airborne platform studies.

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