***Figure S1a.***Mosaic of MODIS “true color” images acquired on Aug. 8, 2013 blinking with their analogue SWIR1 (short wave IR) image (the border of VCT’s Figure 5 image appears as solid red lines on our mosaic). Our true color mosaic shows the presence of orange-coloured forest-fire hotspots over Alaska and the Canadian NWT (North West Territory) region as well as smoke plumes that have made their way from the Great Bear Lake and Great Slave Lake area of the NWT to the open water south of Banks Island (Amundsen Gulf region). The SWIR image shows that the smoke plumes have largely disappeared from view: a signature requirement for FM (fine mode) smoke. The MODIS imagery, MODIS retrievals (of AOD and fine mode fraction), MODIS cloud retrievals (cloud optical depth), CALIOP (attenuated backscatter coefficient and depolarization ratio) profiles, NAAPS smoke modelling and HYSPLIT backtrajectory analyses made it clear (using standard techniques employed, for example, in Ranjbar et al., 2019) that the Banks Island area was generally infiltrated with smoke from the NWT on Aug. 9, 2013, while Aug. 8 smoke from Alaska had made its way to the center of the massive and homogeneous dark blue (negative BTD11-12) region seen in the Aug. 9 image of VCT’s Figure 5. The CALIOP / MODIS AOD products indicated a mean 532 / 550 nm AOD ~ 0.3 for those pixels judged to be aerosols in the region of that dark blue mass on both the August 8 and 9 BTD11-12 images.

1 MODIS bands 7 (SWIR), 2 (NIR) and 1 (R) at 2105-2155, 841-876 and 620-670 nm in the R, G, B display channels. We maintain the name “SWIR” because it arguably has the most impact on the displayed images

***Figure S1b.***True color image of VCT’s Aug. 8, 2013 (Figure 5) blinking with its corresponding (Figure 5) BTD11-12 image. The correlation between the spatial pattern of the smoke plume and the negative (bluish) BTD11-12 values is marginal at best. The labels in quotes are those of the Worldview classification\*

The TIR backscatter of smoke would be largely negligible so that any smoke impact would be due to “Rayleigh” absorption by very optically-small particles whose spectral dependency varies as l-1 (see, for example, Wickramasinghe, 1973). The CALIOP/MODIS 532/550 nm AODs for that smoke event were <~ 0.3 (as per the discussion of Figure S1a). This would result in very weak TIR AODs of <~ 0.03 and correspondingly weak support for any assertion of strongly negative BTD11-12 values being attributable to smoke particles.

\* https://worldview.earthdata.nasa.gov/?v=-162.40677758062284,57.816449317805805,116.30286357841774,82.21310381063934&l=Reference\_Labels\_15m,Reference\_Features\_15m,Coastlines\_15m,OrbitTracks\_Aqua\_Descending,OrbitTracks\_Aqua\_Ascending,MODIS\_Aqua\_Cloud\_Phase\_Infrared\_Day(disabled=4),MODIS\_Aqua\_Sea\_Ice(hidden,disabled=1-2-3-4-5-6-7-8-9-10),MODIS\_Terra\_Cloud\_Optical\_Thickness,MODIS\_Combined\_Value\_Added\_AOD(hidden),MODIS\_Aqua\_Aerosol(hidden),MODIS\_Terra\_Aerosol,MODIS\_Aqua\_AOD\_Deep\_Blue\_Land(hidden),MODIS\_Terra\_AOD\_Deep\_Blue\_Land(hidden),VIIRS\_SNPP\_CorrectedReflectance\_TrueColor(hidden),VIIRS\_NOAA20\_CorrectedReflectance\_TrueColor(hidden),MODIS\_Aqua\_CorrectedReflectance\_TrueColor(hidden),MODIS\_Terra\_CorrectedReflectance\_TrueColor&lg=true&t=2019-08-15-T17%3A44%3A39Z

***Figure S2.*** MODIS-Terra true-colour image acquired at 22:00 UTC, May 29, 2005 blinking with its analogue SWIRimage. The upper half of the MISR frame (the red broken-line rectangle) encloses a more opaque white CM layer or plume (as seen in the blinking SWIR image) that we chose to focus on in terms of the MISR plume height retrievals of Figure S4. This MODIS image shows the western portion of the Canadian Arctic Archipelago from Banks Island in the south to southern Ellesmere Island in the north and the Arctic Ocean to the west of the Archipelago. Coarse mode (CM) particles such as cloud, because of their essentially neutral backscatter spectra, would appear white in both the true colour and SWIR images while snow appears as cyan colored in the latter1.

1 Because of the weak reflectance of snow in the NIR and SWIR; see Schlundt et al., 2011 for example

***Figure S3a.***The BTD11-12 image1 blinking with the SWIR image of Figure S2. The red broken-line rectangle, as discussed in the caption of Figure S2, represents the MISR frame. In blinking mode one can observe negative BTD11-12 (light blue to darker blue) layers that correspond to the white CM layer of the SWIR image. In general the bluish areas are far from any possible local dust sources.

1 With an added corner, to the southwest, taken from the next sequential MODIS image (added for spatial context)

***Figure S3b.***The BTD11-12 image of Figure S3a blinking with the MODIS cloud phase product. The larger amplitude negative BTD11-12 regions generally correspond to “Liquid Cloud” class. The optical depths of the liquid phase clouds are >~ 6 (MODIS “Cloud Optical Thickness” product)

***Figure S4.***MISR nadir image (upon which is superimposed the coloured MISR height retrievals) blinking with the MODIS SWIR image of Figures S2 and S3. The nadir image was acquired at 23:00 UTC on May 29, 20051: its upper 1/3 (corresponding to the region of MISR height retrievals) overlaps the opaque white CM layer seen in the blinking SWIR image (which is truncated to the size of the MISR nadir image). The MISR frame (the red broken-line rectangle whose geographic position is shown on the inset map to the left) corresponds to the MISR frame seen in Figures S2 and S3.

1 One hour later than the MODIS SWIR image. There was no MISR imagery during the orbit line of those figures.

***Figure S5.*** 3D frequency-of-occurrence histograms of MISR-derived plume heights and wind speed for all the height retrieval pixels seen on Figure S4. The plume speed and wind-corrected plume height maximum are substantial and very low-altitude (~ 12 m/s or 42 km/hr and ~ 0.3 – 0.4 km). The 22:00 MODIS-Terra products are largely liquid cloud phase with CODs >~ 6.

***Figure S6.*** NSHSRL (North Slope High Spectral Resolution Lidar) backscatter coefficient (β) profile blinking with NSHSRL depolarization ratio (DR) profile and with the NSA-KAZR radar profile acquired at the Barrow ARM site. The very low DR indicates that the reddish NSHSRL backscatter coefficient profile is likely dominated by a water plume composed of supercooled droplets\* (mixed with ice crystals according to the radar profile; we will continue to refer to a water plume where the DR is very small, even though the total plume is, in general, mixed phase). The optical depth of the water plume is large (see below): in fact it appears to cut off a non-trivial portion of the plume (to the point where CM particle plumes, seen in the radar profile, are not seen in the lidar profile). Note “doubling” of the water plume around 22:00 UT continues on during March 23 until about 03:10 UT when the upper plume appears to disperse. The pink dashed curves represent the hypothesized boundaries of the liquid water plume (the boundaries that were used in the MODTRAN BTD11-12 simulations of Figure 1). That we can hypothesize a water or mixed phase plume above the apparent upper-boundary lidar cutoff is argued below.

\* temperature ~ -20 °C according to the Barrow radiosonde at 12:00 UT

It will be noted that there was no significant temperature inversion across the thin water plume but that there is a significant temperature inversion across the radar-detected plume

Up until about 06:00 UT, the radar profile suggests ice-crystal fallout in the lower 300 m\*\* (that likely results from ice nucleation within the water plume). Fallout events suggestive of even higher altitude CM (radar detectable) sources starting around 08:00 UT are seen in the radar data while being obscured in the lidar data by the near total attenuation of the water plume. Ice clouds around 6 km altitude also occurred in the 1st half of the day (see Figure 1 of the paper).

\*\* which is also captured by the lidar profile (in the apparent absence of liquid droplet backscatter). Ice crystals are also seen in the radar profile above 300 m : but the lidar return due to ice crystals is, in that case, dominated by water plume backscatter

The radar-detected ice clouds are in general, weak (in terms of 532 nm OD) relative to those of the water plume ODs in the visible (and thus the TIR region): the (yellowish) 532 nm NHSRL β value streaks (below the low DR water plume) correspond to what appear to be ice-particle fall-lines \*\*\*. Using that as a rough 532 nm calibration for yellowish radar fall-lines and employing a visible ice-particle lidar ratio ~ 20 sr (see Table 1 of the supplementary information of Ranjbar et al., 2020 for example), the ice crystal visible OD will be ~ 10-5 m-1 sr-1 x 20 sr x 750 m ~ 0.15. This is significantly smaller than the NHSRL (red) water plume OD (~ 10-3.3 m-1 sr-1 x 20 sr x 250 m = 2.5) and so the water plume dominates visible columnar-OD optics (and any point-β optics) in the visible (and the TIR where ODs will be <~ visible OD). The Figure 3 (Dec. 29, 2006) illustration of “liquid” and “mixed-phased” findings of de Boer et al. (2009) at Eureka and the 10-day (May 1-10, 1998) “liquid and “ice” shipborne findings (about 6-7° of latitude north of Barrow) reported by Zuidema et (2005) support a scenario that inversion-layer ice OD can be significantly less than water optical depth.

\*\*\* the first fall-line at ~ 4:40 UT corresponds, for example, to apparent fall-lines of radar β values: the NHSRL β fall-lines are ~ 10-5 m-1 sr-1 versus 10-3.5 m-1 sr-1 = 101..5 x 10-5 m-1 sr-1 = 32 x10-5 m-1 sr-1 for the water plume (32 times the ice crystal β value)

The radar plume segment (layer) that is found above the lidar-detected water plume is in an inversion layer (whereas, as stated above, it is difficult to ascribe a substantial inversion layer to the presence of the red-coloured water plume). That radar layer can represent anything from an optically weak, pure-crystal layer to a mixed phase layer dominated by liquid droplets that would have significant optical depth impact in the visible and TIR but whose liquid droplet size (typically <~ 15 μm radius\*\*\*\*) would largely not be detected by the radar. The negative BTD11-12 results of Figure 1 (supported by the spatial robustness of the BTD11-12 images of Figure S7 and the successful BTD11-12 predictions of Figure 1) argue strongly for the latter being true. The dynamics of liquid water plumes being needed to incite ice nucleation (that fall lines need to be initiated from within a water plume\*\*\*\*\*) also support the latter case.

\*\*\*\* see, for example, the deff statistics of Barrow and Eureka in Figure 8 of de Boer et al. (2009)

\*\*\*\*\* see, for example, Figure 3 of de Boer et al. (2009)

***Figure S7***. BTD11-12 images acquired by the CALIPSO IIR imager on the left and MODIS Aqua on the right. The position of the IIR image is marked by the dashed rectangle on the MODIS Aqua image. The CALIOP (IIR) orbit line is indicated by the purple line. The position of Barrow is marked by a small red “x” on both images and the coastline appears as the black solid curve (at two different spatial resolutions).

***Figure S8***. BTD11-12 vs BT11 simulations for different types of plumes over a “Snow/Ice” surface (surface of Feldman, 2014). (a) and (b): ice or liquid cloud at, respectively a high altitude and within a low altitude temperature inversion layer. (c) and (d): Asian dust plume at, respectively, a high altitude and within a low altitude inversion layer. The general atmospheric conditions and plume parameterization represent a range of values that include the specific conditions of Figures 1a, 1b, S6 and S7 (the specific conditions of Barrow and its surroundings on March 22, 2015). The nominal temperatures employed in the MODTRAN simulations at a (snow/ice) surface, inversion layer plume top and high altitude cloud top were, respectively 255.56, 262.05 and 212.66 °K. The eccentric nature of the low level “Reff = 3 um” red curve in Figure S8d appears to be an approach to a balanced radiative transfer condition wherein there is little change in BTD11with increasing plume optical depth (an approach to an idealistic singularity of a straight vertical line). We determined that this effect was largely due to non linearities in the spectra of the extinction efficiency and the fact that the plume optical depth, τ is fixed, in the MODTRAN simulations, at a single wavelength of 550 nm

The optical properties of the liquid and dust particles were generated with a Mie Code (MiePlot4621, written by Philip Laven (<http://www.philiplaven.com/mieplot.htm>), using the refractive index of water (Hale and Querry, 1973) and dust (Volz, 1973), for monodisperse particles. The optical properties of the ice crystals were extracted from Ping Yang's database (Yang et al., 2013) and correspond to a modified-gamma distribution with effective variance 0.1 (Petty and Huang, 2011) of severely roughened column aggregates (Yang et al., 2013). This is the same distribution that is assumed in the Collection 6 MODIS cloud product (Holz et al., 2016).

For weak DODs associated with high-altitude Asian dust, the BTD11-12 to DOD sensitivity (dBTD11-12 / dDOD) would be best represented by a slope near DOD = 0 (τ = 0 on the graphs) for the case of the near 1.5 μm peak radius of springtime Asian dust (see, for example, the right panel of Figure 16 of Burton et al (2012) and Figure 3 of AeF for the springtime Asian dust particle size distribution). This yields a value (from the detailed numerical results employed in generating these graphs) of -0.30 °K per unit change in DOD).

The brightness temperatures correspond to the EOS-1 TERRA MODIS spectral response functions for bands 31 (max. at 11 µm) and 32 (max. at 11.9 µm), downloaded from this website:

<https://nwp-saf.eumetsat.int/site/software/rttov/download/coefficients/spectral-response-functions/>

A nominal noise figure for MODIS Band 31 (the 11 μm band) is 0.05 °K (the cloud-discrimination ATBD of Ackerman et al., 2010). Given a roughly equivalent (incoherent) noise for band 32 (the 12 μm band) yields a BTD11-12 noise value of √2 ×0.05 = 0.07 °K

***Figure S9.*** True colour (RGB) MODIS-Terra image showing Banks, Prince Patrick and Eglinton Islands (acquired at 22:00 UTC, May 29, 2005) blinking with its analogue BTD11-12 image. The geographic position of those islands within the orange rectangle and relative to the Canadian Arctic Archipelago are shown on the map to the right. For a dust dominated surface (the drainage basin sources of dust on the islands), the dust (“Sand”) “Surface” emissivity spectrum of the Figure 2 table of Vincent (2018) indicates that the values ( values\*) are expected to be strongly negative while the values for the snow and ice regions (according to his “Ice/Snow” “Surface” emissivity spectrum) are expected to be strongly positive. One expects competition between the two signs for the dust deposited on snow (the dark “fingers” of dust deposition extending from the three Islands of the RGB image) and indeed, one can generally see lesser positive values in the neighbourhood of those fingers (with even a few solitary negative pixels in the case of the Prince Patrick Island fingers). That the fingers likely represent dust deposited on snow rather than airborne dust is made clear by their day to day (MODIS image to MODIS image) persistence on the snow (and MISR image sets that show no movement of the plumes between the different MISR cameras). The change in reflectance from the dark areas of the deposited plume and the whitish more pristine areas is ~ 20 to 60%

\* It is easy to show that the of any surface is approximately given by;

where L is the measured radiance and where we ignore the thermal impacts of the plume-free atmosphere. The sign of the image is accordingly driven by .

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