Asian dustfall in the St. Elias Mountains, Yukon, Canada

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Abstract

In April 2001, a major atmospheric dustfall event occurred in the St. Elias Mountains, Yukon Territory, Canada. Field samples were collected and analyzed for particle size, mineralogy, chemical composition and Sr, Nd and Pb isotopes. Dusts found above ~3000 m had their source in the Gobi desert region of northern China and Inner Mongolia, and were transported to the Yukon following a series of major dust storms that took place in early April. Dusts found below 3000 m had local (Yukon) or mixed source(s). The Asian dusts had a modal volume diameter of ~4 µm typical of far-traveled mineral aerosols. However larger (>10 µm) particles were also found at ~5000 m, suggesting a very rapid trans-Pacific transport in the mid-troposphere. We estimate that the April 2001 event deposited from 5500 to 6335 tons of dust over an area of 21,000 km² in the southwestern Yukon, most of which probably fell within a week. Our findings are consistent with instrumental observations and model simulations of the April 2001 event. While the dust cloud was reportedly mixed with volatile pollutants from Asia, we found no evidence of metal pollution associated with the dustfall in the Yukon. Our findings contribute to clarify the dynamics and the geochemical impact of Asian dust long-range transport events, and to better estimate eolian fluxes of dustborne elements (e.g., Fe) to the Ocean associated with such events. They may also assist in identifying past Asian dust events in ice cores drilled from the St. Elias Mountains icefields, to develop a long-term record of their frequency, magnitude and source(s).
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1. Introduction

Every year, 10 to >10² Tg of soil-derived mineral aerosol (hereafter: dust) are transported from the arid East Asian interior to the North Pacific atmosphere, some of it reaching North America (McKendry et al., 2001; Vaughan et al., 2001; VanCuren and Cahill, 2002; Zhao et al., 2003; DeBell et al., 2004). In addition to their adverse effects on air quality and visibility near the source regions (China, Mongolia, Korea), these dust long-range transport (LRT) events can affect the atmosphere's com-

position and radiative balance on an hemispheric scale (e.g., Seinfeld et al., 2004). They can also deposit large amounts of mineral nutrients (e.g., Fe) in the Ocean, impacting surface biological productivity and the air–sea exchange of CO₂ (Bishop et al., 2002; Johnson et al., 2003), and can act as carriers of air pollutants (e.g., Nishikawa et al., 1991). Asian dust can be detected almost year-round in the North American atmosphere (VanCuren and Cahill, 2002) and has been found in snowpacks as far as Greenland (Biscaye et al., 1997; Svensson et al., 2000; Bory et al., 2002, 2003a,b) and the French Alps (Grousset et al., 2003). The frequency and magnitude of East Asian dust storms may now be increasing as a result of climate warming, droughts and spreading desertification (United Nations Environmental Programme, 2004). Hence, there is a need to better

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understand and characterize the impact that future Asian dust outbreaks may have across the North Pacific region.

The LRT of Asian dust to Canada was previously observed in the Northwest Territories (Welch et al., 1991) and British Columbia (McKendry et al., 2001). Here, we report on a more recent episode that affected the southern Yukon Territory. Evidence of this LRT event was found in early May 2001 by members of a glaciological expedition, who observed a widespread layer of dust in the icefields of the central St. Elias Mountains. This finding was tentatively linked to major Asian dust storms that had taken place in April of that year (Wang et al., 2002). In this paper, we investigate the provenance of the dusts found in the St. Elias Mountains by comparing their physico-chemical attributes with those of Asian and proximal (Yukon) sources. In doing so, we evaluate the reliability of mineralogical, chemical and isotopic properties for discriminating between contrasted dust sources. We also estimate the mass of dust deposited in the St. Elias Mountains and its geochemical impact, and we compare our findings with other measurements and model simulations of the well-documented April 2001 LRT event.

2. Geographic and climatological setting

The study area is located in the central St. Elias Mountains, southwestern Yukon Territory (Fig. 1). The area numbers 13 peaks >3000 m, including Mt. Logan, Canada's highest mountain (5959 m). The peaks are surrounded by large icefields and glaciers that flow into the Yukon interior or towards coastal Alaska. The St. Elias and nearby Wrangell Mountains form a major orographic obstacle to westerly airflow and moisture from the North Pacific. As a result, the area is characterized by abundant precipitation on the Alaskan slopes, and a semi-arid climate in the Yukon interior.

The climatology of the April 2001 dust storms was extensively described elsewhere and is only summarized here (Wang et al., 2002; Jaffe et al., 2003a; Wu et al., 2003; Uno et al., 2004). On April 6–7, 2001, a deep cyclonic depression associated with a vigorous cold front lifted huge amounts of dust across a wide area of northwestern China and Inner Mongolia. A second depression followed on April 8–9, causing further dust outbreaks in northeastern and north-central China. The resulting massive dust cloud moved off the Asian landmass and was rapidly transported across the North Pacific Ocean to reach western Canada on April 12 and the Gulf of Alaska ca April 14 (Thulasiraman et al., 2002; Cahill, 2003). Haze caused by the dust was observed in Anchorage on April 16, coincident with a sharp increase in soil-derived aerosols measured at the Poker Flats Research Range, ~30 km northeast of Fairbanks (Cahill, 2003). The timing matches observations made by the Interagency Monitoring PROgram for improved Visual Environments (IMPROVE) in the northwestern USA (Jaffe et al., 2003b). Measurements of the aerosol index

Fig. 1. (a) Map of the southwestern Yukon Territory showing geographical features mentioned in the text. (b) Sites in the Mt. Logan area from where dust samples were obtained.

(an indicator of UV-absorbing particles) by NASA's spaceborne Total Ozone Mapping Spectrometer (TOMS; <http://toms.gsfc.nasa.gov>) show that dust levels over southern Alaska and the Yukon were highest between April 14 and 18. Air mass back-trajectories computed with NOAA's web-enabled HYSPLIT4 (Hybrid Single-Particle Lagrangian Integrated Trajectory) model suggest that Asian dust reaching Mt. Logan during this period was initially transported in the mid-troposphere (>700 hPa or >3000 m) and subsided in the lower troposphere in the lee of a cold front moving over the region ca April 12 (Fig. 2). This is consistent with aerosol scattering measurements in the northeastern Pacific which showed that the dust was mostly transported above 3000 m (Price et al., 2003). The duration of the dustfall period in the St. Elias Mountains could not be ascertained owing to lack of direct observations. However, a period of intensified precipitation (rain and snow) was reported in Yakutat, coastal Alaska, and in the Yukon interior between April 11 and 19, 2001 (Alaska Weather Bureau, Environment Canada). Our own observations suggest that dustfall in the Mt. Logan area was accompanied and followed by snowfall. Most of the deposition may therefore have occurred during this ~1-week period.

Fig. 2. Modeled 5-day back-trajectories for air masses reaching Mt. Logan (star) on April 16, 2001. The end-points are at Prospector-Russell Col (PRC; 5340 m), King Col (KC; 4150 m) and on the Quintino-Sella Glacier (QS; 2800 m). Trajectories were computed with the HYSPLIT 4 model of the NOAA Air Research Laboratory. Also shown are frequent source areas of Asian dust storms: TD, Taklimakan desert, China; GD, Gobi desert, China/Mongolia; LP, Loess Plateau, China.

Dust flux measurements in China (Zhang et al., 2003a) and satellite observations (TOMS) showed that the April 2001 dust cloud was primarily emitted from sources in the Taklimakan desert (Tarim Basin), and in north-central China and Inner Mongolia (Gobi desert). However different model simulations of the event disagree on which of these regions contributed most of the dust transported across the North Pacific (Gong et al., 2003; Uno et al., 2004). The data presented here may help to clarify the dust transport history for this event.

3. Methods

3.1. Sample collection

3.1.1. Dust samples

Snow samples mixed with dust were collected in May and June 2001 from sites on or near the Mt. Logan massif (Fig. 1, Table 1). On the Quintino-Sella and Seward glaciers (2000–2800 m), the dust was found in a ~1–3 cm thin, patchy, reddish-brown snow layer. Higher up in King Trench (2800–4000 m), at King Col (4150 m) and at Prospector-Russell Col (5340 m) the dust was found in a 5–12 cm thick brown snow layer. This layer was more compact than the snow above or below it, suggesting relatively moist and/or turbulent conditions when the dust fell. A separate dust sample was also collected in June 2002 from a snowpit excavated near Mt. Queen Mary, ~40 km east of Mt. Logan (Fig. 1). The sample was taken in a 5-cm thick layer of dirty snow found at a depth of 275 cm. On strati-

graphic grounds, it was estimated to have been deposited in the spring of 2001, and was therefore attributed to the same dustfall event sampled near Mt. Logan. While sampling, care was taken to avoid contaminating the snow with clothing or dirty tools. Clean plastic scoops were used to pour snow into clean HDPE bottles or sealable LDPE bags. All samples were shipped and stored in frozen state until prior to analysis. In the text, we identify the dust samples by referring to the sites from which they were taken, using a simple letter code (Table 1), for example: KTU, Upper King Trench; MQM, Mount Queen Mary.

3.1.2. Local (Yukon) dust sources

Some of the dusts deposited in snows of the St. Elias Mountains may have proximal sources. Outwash flats from the glacier-fed Slims and Donjek rivers (Fig. 1) are the site of frequent local wind storms in the spring and summer, which entrain and disperse dust clouds over a broad area (Nickling, 1978). Widespread loess deposits of late Pleistocene to Neoglacial age found near Kluane Lake were probably formed by this process (Denton and Stuiver, 1966). In order to discriminate between exotic (Asian) and local (Yukon) sources, we compared the composition of the dusts found in the St. Elias icefields with that of loess collected in the Kluane Lake area. We assume that the loess is an acceptable surrogate for eolian dust produced locally under present conditions. In this paper we limit our discussion of the Yukon loess properties as they pertain to dust source discrimination. Details on the loess sampling sites and a more complete discussion of analytical results will be presented elsewhere.

3.2. Analytical

To identify the source(s) of dusts sampled in the St. Elias Mountains, we considered various source-specific properties that can be conserved during long-range windborne transport. These include clay mineralogy, major or minor elemental ratios, rare earth element (REE) abundances, and Sr, Nd and Pb isotope ratios (Schüetz, 1989; Zhang et al., 1996; Biscaye et al., 1997; Grousset and Biscaye, 2005). We also analyzed the particle size distribution of the dusts, which is related to transport distance. A subset of these properties were analyzed in the <5 µm size fraction of the Yukon loess samples, following nearly identical methods (described below). We chose this particle size range because it is typical of dusts transported over long distances ($\geq 10^2$ km; Schüetz, 1989), and also ensures comparability of our loess data with Asian desert dusts analyzed by Biscaye et al. (1997). The amount of dust in our snow samples varied between 5 and >900 mg, and not every sample could be analyzed for all properties. Whenever possible, samples were divided to allow for multiple analyses (Table 1). We extracted the dust by filtration or evaporation of the melted snow, except for the large sample collected at site MQM which was freeze-dried to check for the presence of water-soluble carbonates in the dust.

The particle size distributions in four dust samples (sites PRC, KTU, QSA and MQM) were determined by laser light scattering using a Leco-trac™ LT-100 Particle Size Analyzer (PSA), following procedures outlined by Girard et al. (2003). The detection sensitivity of the PSA dropped rapidly for sub-micron particles. Hence, we report data for particles >1 µm only.

Two dust samples were analyzed by powder X-ray diffraction (XRD) to determine their mineralogical composition. The sample from site KC

Table 1
List of dust samples analyzed and methods employed in this study

Sampling location ^a	Code	Lat. (N)	Long. (W)	Elev. (m)	Date sampled (m/d/y)	Weight of (sub-) sample		Dust extraction method	Analyses performed	Method(s) ^c
						Snow (g)	Dust ^b (mg)			
Prospector-Russel Col	PRC	60° 36'	140° 30'	5340	6/19/01	196	n.a.	No extraction	Part. size distribution	PSA
King Col	KC	60° 35'	140° 36'	4150	5/25/01	924	16.1	Melting and evaporation	Sr and Nd isotopes Mineralogy	TIMS XRD
Upper King Trench	KTU	60° 36'	140° 39'	3800	5/28/01	307	24.1	Filtration	Geochemistry	ICP-ES, ICP-MS
						299	8.0	Filtration	Geochemistry	ICP-ES, ICP-MS
						311	n.a.		Part. size distribution	PSA
Lower King Trench	KTL	60° 36'	140° 42'	2950	6/30/01	256	20.0	Filtration	Geochemistry	ICP-ES, ICP-MS
						322	5.6	Filtration	Geochemistry	ICP-ES, ICP-MS
Quintino-Sella Glacier	QSA	60° 36'	140° 49'	2800	6/10/01	262	n.a.	Filtration	Part. size distribution	PSA
	QSB	60° 36'	140° 49'	2800	6/10/01	234	16.9	Filtration	Geochemistry	ICP-ES, ICP-MS
						n.a.	27.0	Filtration	Sr and Nd isotopes	TIMS/MC-ICP-MS
	QSC	60° 34'	140° 58'	2460	6/1/01	2518	37.0	Filtration	Geochemistry	ICP-ES, ICP-MS
Seward Glacier	SG	60° 26'	140° 16'	2000	6/27/01	316	25.4	Filtration	Geochemistry	ICP-ES, ICP-MS
						521	26.4	Filtration	Geochemistry	ICP-ES, ICP-MS
Mount Queen Mary	MQM	60° 41'	139° 47'	2620	6/25/02	330	521.0	Freeze-drying	Part. size distribution	PSA
						304	532.0	Freeze-drying	Geochemistry	ICP-ES
						349	438.0	Freeze-drying	Pb, Sr and Nd isotopes	TIMS/MC-ICP-MS
									Mineralogy	XRD
						352	906.0	Filtration	Geochemistry	ICP-ES, ICP-MS

n.a., data not available.

^a See Fig. 1.

^b Weight of solid particulates (>0.45 µm) extracted from the snow.

^c PSA, laser scattering particle size analyzer; XRD, X-ray powder diffractometer; ICP-ES (MS), inductively coupled plasma emission (mass) spectrometer; MC-ICP-MS, multi-collector ICP-MS; TIMS, thermal ionization mass spectrometer.

was analyzed on a Phillips X'Pert™ multi-purpose diffractometer at the Lamont-Doherty Earth Observatory (LDEO) using the procedures of Biscaye et al. (1997). Relative percentages of chlorite, illite and kaolinite + chlorite were estimated from the area of the chlorite 14 Å (001), illite 10 Å (001) and kaolinite (001) + chlorite (002) 7 Å peaks as described in Biscaye (1964, 1965). The coefficient of variation for the calculated mineral percentages is conservatively estimated to be ±10%. The dust from site MQM was analyzed at the Geological Survey of Canada (GSC) on a Bruker D8 Powder diffractometer with a monochromatic Co Kα radiation source ($\lambda = 1.79$ Å), and relative mineral weight percentages ($\geq 1\%$) were estimated. The same method was used for the Yukon loess samples. Since no internal standards were used, errors on the mineral percentages could not be estimated, but could be as high as ±20–25% (Percival et al., 2001).

The concentrations ([x]) of 45 major, minor and trace elements were determined in the dust and snow after filtration of the melted samples through 0.45 µm Durapore filter membranes in a class 1000 environment. The filtered water was acidified with HNO₃ and analyzed using the methods described in Hall et al. (1995, 1996). The filters were air-dried, weighed, and the particulate material was dissolved in Teflon beakers using a gently heated mixture of HF and aqua regia (HCl + HNO₃). After dissolving the dust, the solution was evaporated to near-dryness and the residue was re-dissolved in aqua regia and diluted to 20 mL in HDPE bottles prior to analysis. Major element analyses were performed on a Perkin-Elmer 3000DV Inductively Coupled Emission Spectrometer (ICP-ES) with a cross-flow nebulizer, while minor and trace elements were

analyzed on a VG PQII Mass Spectrometer (ICP-MS) with a concentric glass nebulizer. The same analytical procedure was used for the Yukon loess samples. Full results are given in annexes EA-1 (dust samples) and EA-2 (Yukon loess). Because the dust and loess samples were acid-digested prior to analysis, we do not report [Si].

Isotopic ratios of Sr (⁸⁷Sr/⁸⁶Sr), Nd (¹⁴³Nd/¹⁴⁴Nd) and Pb (²⁰⁷Pb/²⁰⁴Pb and ²⁰⁶Pb/²⁰⁴Pb) were determined in three dust samples with a Thermal Ionization Mass Spectrometer (TIMS) and/or a Multiple-Collector Inductively Coupled Mass Spectrometer (MC-ICP-MS). The dust from site KC was analyzed at LDEO by TIMS for Sr and Nd isotopes only, following methods described by Svensson (1998) and Bory et al. (2003b). The XRD trace for this sample showed no detectable carbonate minerals. Hence, Sr and Nd isotopic ratios are considered representative of the silicate mineral fraction. Dusts from sites QSB and MQM were analyzed for Sr, Nd and Pb isotopic ratios, and for [Sm] and [Nd] at the GSC's Tracer Isotope Laboratory using a Nu-Plasma MC-ICP-MS and/or a Triton-TI TIMS. Isotopic ratios were determined by MC-ICP-MS and TIMS for Sr, and by TIMS for Pb, while [Sm] and [Nd] were measured by MC-ICP-MS. The Sr isotope ratios measured by TIMS and MC-ICP-MS are consistent within error, and we report weighted means of TIMS and MC-ICP-MS measurements as the representative values. Sample digestion and chemical separation methods are described by Amelin et al. (1997) and Amelin and Zaitsev (2002). The dust from QSB was filtered from meltwater, whereas that from MQM was freeze-dried. The XRD trace of the dust from MQM showed 1–3% (weight) of calcite and dolomite, but the presence of carbonates in the sample from QSB could not be verified owing to limited sample volume. Because the

$^{87}\text{Sr}/^{86}\text{Sr}$ ratio in carbonates can differ significantly from that in silicates, a split from each sample was partially dissolved in dilute acetic acid (HOAc) and the residue was analyzed separately. An identical procedure was used for the Yukon loess samples. Full results are given in [annex EA-3](#). The measured $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were corrected for mass discrimination using exponential normalization to $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$. Results are expressed relative to a mean value of $^{87}\text{Sr}/^{86}\text{Sr} = 0.71025$ in NIST standard SRM-987. The $^{143}\text{Nd}/^{144}\text{Nd}$ ratios were corrected for spike contribution and mass discrimination by numerically solving isotope dilution equations through exponential normalization. Quality control was performed by monitoring the uniformity of non-radiogenic isotopic ratios ($^{145}\text{Nd}/^{144}\text{Nd}$, $^{150}\text{Nd}/^{144}\text{Nd}$, $^{150}\text{Sm}/^{152}\text{Sm}$ and $^{154}\text{Sm}/^{152}\text{Sm}$) and by analysis of standards (La Jolla Nd and Ames Sm). The Nd isotope ratios are reported here in the epsilon notation, where $\epsilon\text{Nd}_{(0)}$ are departures ($\times 10^{-4}$) of the $^{143}\text{Nd}/^{144}\text{Nd}$ ratio from the present-day bulk Earth value of 0.512638 (Jacobsen and Wasserburg, 1984). For the Pb isotope ratios, mass-discrimination effects were corrected using average isotopic ratios measured in the NIST “common lead” standard SRM-981 and in the standard rock BCR-1, determined with double isotope dilution (Woodhead and Hergt, 2000). Both methods gave average mass discrimination values that agree at the limits of their 2σ errors.

4. Dust properties and provenance

4.1. Particle size distribution

Particle size distributions of the dusts found at sites PRC, KTU, QSA and MQM are shown on [Fig. 3](#) by number (N) and volume (V). All four samples have a common number mode $\bar{D}_N \approx 2 \mu\text{m}$, and three (PRC, KTU and QSA) also share a volumetric mode $\bar{D}_V \approx 4 \mu\text{m}$. This mode is typical of far-traveled dusts deposited in alpine snowfields (Wagenbach and Geiss, 1989; Wake et al., 1994; Osa-da et al., 2004), and is therefore consistent with a distant source. It is also close to the volume-weighted modal radius of $1.8 \mu\text{m}$ for particles in the April 2001 dust cloud inferred by Thulasiraman et al. (2002) from sunphotometric observations. However, PSA results also reveal the presence of large ($>10 \mu\text{m}$) particles in dusts deposited at sites MQM and PRC. At MQM these particles are sufficiently abundant ($\sim 10\%$ by numbers) to skew the entire volumetric distribution to a mode $\approx 70 \mu\text{m}$.

The distinctly coarse particle size distributions observed at MQM and PRC demand separate interpretations. The dust from MQM was sampled in the accumulation area of the Kaskawulsh Glacier ([Fig. 1](#)). Although it was first attributed to the April 2001 dustfall event, its coarseness suggests instead a proximal source, possibly in the nearby Slims River valley. The dust sample from PRC (Prospector-Russell Col; 5340 m) may contain particles eroded from exposed rocks outcrops. However the sampling site at KTU (Upper King Trench; 3800 m) was much closer to such outcrops and yet the dust collected there shows no comparably coarse particles. We therefore suggest that the large dust particles detected in snow at PRC are of Asian origin, and were transported at this altitude by fast-moving mid-tropospheric winds. This interpretation is consistent with optical properties of the April 2001 dust cloud which indicated the presence of such large particles in it (Chung et al., 2003; Price et al., 2003). The absence or scarcity of comparable particles in the dusts found at

QSA and KTU suggests that they were possibly deposited at altitudes $\geq 4000 \text{ m}$, where most of the dust mass was transported (Price et al., 2003). Unfortunately the dust sample from PRC was too small to verify its provenance by compositional criteria, and our interpretation of PSA results therefore remains tentative.

4.2. Mineralogy

The XRD analyses performed on the dust sampled at site KC on Mt. Logan showed that it is composed of quartz and feldspars (65% weight), mica and/or illite (21%), chlorite and kaolinite, with traces of smectite or other mixed-layer clay minerals. This assemblage is fairly typical of Asian dust (e.g., Leinen et al., 1994; Kanayama et al., 2002), but the high proportion of quartz and feldspars suggests the presence of some silt-size or coarser particles in the dust, as confirmed by PSA results from other samples. Because windborne clay-size dust particles have low settling velocities, their mineralogy is more likely to resemble that of their parent soil. As shown in [Table 2](#), the clay mineralogy of the dust found at KC is indeed much closer to that of Chinese loess than of Yukon loess, which confirms its exotic origin. The kaolinite to chlorite ratio of the dust is also within the narrow range of ~ 0.4 – 0.5 characteristic of northern Chinese desert dusts (Biscaye et al., 1997; Sun et al., 2000). The dust found at MQM differs by the presence of dolomite ($\sim 13\%$ weight), and the lack of detectable kaolinite. In this regard it is compositionally closer to the Yukon loess, which supports a local source for the dust in this sample, as its coarseness suggested.

4.3. Major and minor element ratios

We compared the proportions of selected elements (Al, Ca, Fe, K and Mg) analyzed in the dusts found in the St. Elias Mountains with those in Yukon loess ([Fig. 4](#)), and in Chinese desert dusts sampled at or near their source, or transported across the North Pacific Ocean ([Fig. 5](#)). Our choice of elements to consider was limited by the availability of published data. We stress that such comparisons can only highlight major similarities or differences between our samples and published aerosol data, because of the diversity of analytical methods used, and of particle size ranges considered. Also, we did not compare our dust samples directly with Asian desert soils because elemental ratios can be modified from soil to dust due to wind sorting of particles (e.g., Rahn, 1976).

[Fig. 4](#) shows that dusts sampled at four sites on Mt. Logan (QSA, QSC, KTL and KTU) are chemically very similar, and clearly distinct from the Yukon loess. Their major element ratios are close to those in the mean upper continental crust (UCC), as can be expected in far-travelled, well-mixed eolian dusts (Taylor et al., 1983). These dusts are therefore presumed to have a common, distant Asian source. In contrast, the dusts sampled at sites QSB, SG and MQM have compositions near or within the com-

Fig. 3. Particle size distributions of dusts collected in the snows of the St. Elias Mountains, Yukon. The distributions are shown as relative and cumulative number (N) and volume (V) frequency curves plotted against particle diameter D . Each curve is an average of three replicate analyses. The shaded area for the sample taken at site PRC shows the range of results for individual analyses; the variance for replicate analyses of samples taken at sites KTU, QSA and MQM was comparatively negligible. The number ($\bar{D}_N$) and volumetric ($\bar{D}_V$) mode(s) of each size distribution are indicated.

positional range of Yukon loess, indicating that they were probably derived, in whole or in part, from local sources. At the four sites where Asian dust was sampled, most of the chemical burden resided in the particulates ($>45 \mu\text{m}$; annex EA-1). This is true for most elements with the exception of Ca, S and Sr, for which 63%, 61%, and 33% (average) of the total burden, respectively, was measured in the filtered meltwater. Some of these elements may have a marine (sea salt) component mixed with crustal material (Ma et al., 2004). We estimated maximum sea-salt contributions of Ca, S and Sr using seawater ratios (Keene et al., 1986),

assuming all water-soluble Na in the snow is from sea spray. The estimated ratios of sea-salt to non-sea-salt fractions were $\leq 3\%$ of total concentrations, implying that most of the Ca, S and Sr came from the dust itself, probably in the form of salts, sulphates or carbonates which are common in arid soils of northern China (Makra et al., 2002; Okada and Kai, 2004; Wang et al., 2004a, 2005).

Hinkley (1994) measured rock-forming metals of the alkali (K, Rb and Cs) and alkaline earth (Ca, Sr and Ba) groups in snows of the St. Elias Mountains in south-eastern Alaska. Snow layers deposited in April–May had

Table 2

Clay mineralogy of samples analyzed in this study, compared to Asian desert dusts and North Pacific clays

Sample type/location	Size (μm)	N	Clay mineralogy ^a (relative weight %)				Kaolinite/chlorite	Reference
			Mica and/or Illite	Chlorite	Kaolinite	Smectite		
<i>Dust in snow, St. Elias Mountains</i>								
KC	Bulk	1	Mean = 60	27	12	1	0.39 ^b	This study
MQM	Bulk	1	Mean = 71	27	0	<1	<0.002 ^b	This study
<i>Yukon dust sources</i>								
Loess (Kluane Lake area)	<5	5	Mean = 43 Range = [31–55]	56 [45–69]	0 n.a.	<1 n.a.	<0.003 ^b	This study
<i>Asian dust sources</i>								
Loess ^d (China)	n.a.	20	Mean = 60 Range = [49–72]	7 [3–10]	22 [17–29]	10 [1–17]	3.14 ^c	Liu (1988)
Loess (China)	<5	3	Mean = 58 range = [56–59]	27 [26–28]	15 [10–17]	Trace n.a.	0.51 ^c [0.35–0.65]	Biscaye et al. (1997)
Gobi desert dust (Mongolia)	<5	3	Mean = 41 Range = [28–60]	39 [30–48]	20 [10–25]	Trace n.a.	0.51 ^c [0.34–0.66]	Leinen et al. (1994)
<i>Asian mineral aerosols</i>								
China (Gobi source)	<5	1	Mean = n.a.	n.a.	n.a.	n.a.	0.45 ^b	Sun et al. (2000)
North Pacific Ocean	<2	46	Mean = 76 Range = [48–89]	6 [2–10]	19 [0–44]	3 [0–22]	3.17 ^c	Leinen et al. (1994) Arnold et al. (1998)
	2–20	42	Mean = 77 Range = [50–89]	9 [2–24]	15 [0–32]	5 [2–12]	1.67 ^c	Leinen et al. (1994) Arnold et al. (1998)
<i>Deep-sea pelagic clays^e</i>								
North Pacific Ocean	<2	n.a.	Range = [30–40]	[10–15]	[10–15]	[0–5]	n.a.	Leinen et al. (1989)

n.a., data not available.

^a As determined by X-ray diffraction (XRD) analysis, unless otherwise indicated.^b Calculated from XRD peak area ratio.^c Calculated from mean weight percentages as determined by XRD.^d Mineral percentages determined by petrographic method.^e May include hemipelagic clays of non-eolian origin.

the highest metal concentrations, and their K/Ca, K/Cs, K/Rb and Ca/Ba ratios showed that these metals were associated with unwinnowed (clay-poor) dusts from relatively proximal and/or unweathered terranes of ferromagnesian composition. Rocks exposed on southwest-facing slopes of the Mt. Logan massif, where most of our samples were collected (Fig. 1), belong to the metasedimentary and metavolcanic Valdez Group and Seward Suite, which are predominantly ferromagnesian (biotite- and hornblende-rich; Israël, 2004). Yet the snow layers sampled from sites QSA, QSC, KTL and KTU have much lower K/Cs (2994 ± 860) and Ca/Ba (29 ± 6) ratios, and a higher K/Ca ratio (2 ± 1) than those measured by Hinkley (1994) in Alaska (K/Cs = 4685 ± 757 ; Ca/Ba = 171 ± 16 ; K/Ca < 1). These differences indicate that most of the dusts found at these sites were not derived from local rocks. Furthermore, relative proportions of Al, Fe, K and Mg in these dusts closely resemble those in mineral aerosols ascribed to sources in northern China or Mongolia (Fig. 5). They are also remarkably similar to Gobi desert dusts transported to the USA and Canada in April 1998 (McKendry et al., 2001; Vaughan et al., 2001). However their Ca/Al and Ca/Fe ratios are noticeably lower than in most Asian dusts sampled near their source(s). This is also true of dusts sampled in April 2001 by the

IMPROVE (northwestern USA; Jaffe et al., 2003b) and AIRMAP networks (northeastern USA; DeBell et al., 2004), and, to a lesser extent, of the dust from the April 1998 event. Mori et al. (2003) showed that the Ca/Al ratio of dust transported from China to Japan tends to increase during transit as aluminosilicate particles settle out. Other processes must therefore be invoked to explain the lower Ca/Al (and Ca/Fe) ratios in the trans-Pacific Asian dusts. These processes may include precipitation scavenging and dissolution of Ca-bearing minerals (e.g., CaSO_4 ; Ma et al., 2004), or heterogeneous phase reactions with acidic aerosols (Krueger et al., 2004). Whatever the process(es) involved, our findings suggest that elemental ratios to Ca in airborne dust are not conserved during LRT, and are therefore unreliable as source tracers. However, the ratios of other elements such as Al, Fe and K can apparently discriminate between sources in northwestern and north-central Chinese deserts, and these point to the Gobi desert as the main source region for the dust deposited in the St. Elias Mountains in April 2001.

4.4. Rare earth elements

The relative abundance of the lithophile rare earth elements (REE) is nearly uniform in loess deposits world-

Fig. 4. Major element chemistry (Al, Ca, Fe, Mg and K) of dusts sampled in the snows of the St. Elias Mountains, compared with the compositional range for Yukon loess. The dust samples are plotted using elemental abundances in particulates ($>0.45 \mu\text{m}$ filtered solids; full circles) and in snow (filtered solids + meltwater; open circles). UCC, mean upper continental crust (Taylor and McLennan, 1985).

wide (Gallet et al., 1998). Minor departures from this pattern, however, may be source-specific (e.g., Cullers et al., 1987). Fig. 6 shows chondrite-normalized REE patterns ($[\text{REE}]_N$) in the dusts sampled on or near Mt. Logan in May–June 2001. Dusts with a probable Asian source (from sites QSA, QSC, KTL and KTU) show typical loess-like patterns, with the lighter REE (La–Sm) enriched relative to heavier ones (Gd–Lu), and a negative europium anomaly ($\text{Eu}_N/\text{Eu}^* \approx 0.65$). The $[\text{REE}]_N$ patterns in these dusts are distinct from those in Yukon loess, but very close to those of Chinese loess and fine desert soils (Ding et al., 2001; Jahn et al., 2001; Honda et al., 2004; Yokoo et al., 2004), and identical to those of Asian dusts found in Greenland ice (Svensson, 1998). While also loess-like, the dusts found at sites QSB, SG and MQM differ in several ways. In particular, the dust from SG has much higher $[\text{REE}]$, hence lower $[\text{REE}]_N$ than all other samples (Fig. 6). Other differences are revealed by comparing REE ratios (Gd/Yb, La/Sm, and La/Lu) and Eu anomalies (Fig. 7). In these plots the dusts from QSB and MQM clearly stand out as distinct from other samples. The dust from QSB is somewhat closer to the Yukon loess and is therefore likely in part of local origin. The sample from MQM has the

highest La/Lu ratio (161.1) and smallest Eu anomaly ($\text{Eu}_N/\text{Eu}^* \approx 1$), features which are compatible with a high plagioclase content in this sample (58% weight), as feldspars are typically Eu-enriched and also have high light-to-heavy REE ratios (Taylor and McLennan, 1981). These findings, and the coarseness of the dust found at MQM, also argue for a proximal source and short transport distance. In contrast, the dusts from sites QSA, QSC, KTL and KTU have REE characteristics that are much closer to Chinese desert dust sources (Fig. 7). In particular, their Gd/Yb and La/Sm ratios closely resemble those in loess from the Loess Plateau (LP) in north-central China. This is consistent with a source for these dusts in the Gobi desert region of northern China and Mongolia, which is also thought to be the source of the LP loess (Sun, 2002).

4.5. Sr, Nd and Pb isotopes

On Fig. 8 we compare the Sr, Nd and Pb isotopic ratios of dusts sampled in the St. Elias Mountains (sites KC, QSB and MQM) with those in Yukon loess, in Asian dusts deposited in Greenland ice (Biscaye et al., 1997; Svensson, 1998; Bory et al., 2003a), and in Chinese loess and desert

Fig. 5. As in this figure, but compared with aerosol data from the April 2001 event and from previous Asian dust storms sampled at or near their source. Dust samples of suspected local (Yukon) origin were not included. Aerosol data from the northwestern USA: IMPROVE network (Jaffe et al., 2003); northeastern USA: AIRMAP network (DeBell et al., 2003); April 1998 trans-Pacific dust event: McKendry et al. (2001), Vaughan et al. (2001); Taklimakan desert dusts: Zhang et al. (1996), Zhou et al. (1996), Makra et al. (2002); Gobi desert dusts: Parungo et al. (1994), Mori et al. (2003), Zhang et al. (1993, 2003a). Also shown is the mean dust composition from northern and northwestern high-dust deserts in China (Zhang et al., 2003b).

soils (Biscaye et al., 1997; Jones et al., 2000; Bory et al., 2003a; Nakano et al., 2004). For these comparisons we used isotopic ratios determined from carbonate-free material only, which can be considered representative of the silicate fraction (annex EA-3 and published data).

The Sr and Nd isotope ratios of the dust found at site KC on Mt. Logan are undistinguishable from those of Asian dusts deposited in central Greenland, and very close to those in desert dusts from northern China and/or southern Mongolia. In contrast, the dust found at site MQM has Sr, Nd and Pb isotopic ratios close or identical to those in Yukon loess, which further confirms its local origin. The dust sampled at site QSB has an intermediate composition between Asian and local (Yukon) sources, indicating a mixture of both. A simple binary mixing model suggests that it contains 72% Asian dust and 28% local dust. These results confirm that the April 2001 Asian dustfall episode in the St. Elias Mountains left its most distinctive imprint at elevations >3000 m, where most of the dust was transported (Price et al., 2003). Of all properties investigated, the isotopic ratios of Sr, Nd and Pb also prove to be the most capable of discriminating between Asian and local (Yukon) sources.

5. Discussion

To quantify the April 2001 dustfall in the St. Elias Mountains, we estimated the mass of dust (M_{dust}) deposited over the area bounded by the British Columbia and Alaska borders, and by the Shakhwak Trench to the east (Fig. 1). A digital elevation model was used to calculate the hypsometry of the area ($\approx 21,000 \text{ km}^2$). We estimated M_{dust} by extrapolating dust concentrations in snow over the whole area, assuming linear relationships between snowfall rate and elevation (Keeler, 1969; Marcus and Ragle, 1970). In the absence of direct observations of the dustfall event, we considered two possible depositional scenarios, one involving snow scavenging only (1), the other dry fallout only (2). In (1), the dust concentration in snow was assumed to increase with snowfall, while in (2) a constant value was used. As discussed earlier, there are grounds to believe that snow scavenging was the dominant mechanism of dust deposition. Depending on the scenario (1 or 2), our estimates of M_{dust} vary from 5500 to 6335 tons and a realistic figure may lie between. Assuming the dustfall lasted ~ 7 days, the mean dust flux may have ranged from ~ 40 to $45 \text{ mg m}^{-2} \text{ day}^{-1}$. These

Fig. 6. Chondrite-normalized REE patterns for dusts sampled in the snows of the St. Elias Mountains, Yukon, compared with patterns (shaded) measured in (a) Yukon loess (this study), (b) Asian dust deposited in Greenland ice, (c) Taklimakan desert dust, northwestern China; and (d) loess and desert soils from the Loess Plateau, north-central China. The chondritic composition used for normalization is that of Anders and Grevesse (1989). Greenland data are from Svensson (1998); Taklimakan desert: Honda et al. (2004), Loess Plateau: Ding et al. (2001), Jahn et al. (2001), Honda et al. (2004) and Yokoo et al. (2004).

calculations are of course simplistic, as neither dustfall nor snowfall were spatially uniform. However they allow for some interesting comparisons to be made against model simulations and instrumental observations of the April 2001 event.

For example, simulations with the Northern Aerosol Regional Climate Model (NARCM) suggest that atmospheric dust loadings (>700 hPa level) over the southern Yukon during the April 2001 LRT event ranged between 50 and 200 mg m^{-2} (Gong et al., 2003; Zhao et al., 2003). If dustfall in the central St. Elias Mountains lasted ~ 1 week, and the daily mean dust loading during this period was $\sim 100 \text{ mg m}^{-2}$ (Gong et al., 2003), up to $\sim 45\%$ of it must have been deposited to match our own dust flux estimates. Coincidentally, Cahill (2003) found that the mass of coarse ($1.15\text{--}2.5 \mu\text{m}$) and fine ($0.34\text{--}1.14 \mu\text{m}$) dust aerosols transported from the Aleutian Islands (April 12, 2001) to the Alaskan interior (April 16, 2001) dropped by 85%

Fig. 7. REE ratios in dusts from the St. Elias Mountains compared with those in Yukon loess ($<5 \mu\text{m}$, this study), Asian dust in Greenland ice [1, shaded], and potential Chinese desert sources [2–4]. (a) Eu_N/Eu^* vs La/Lu ; (b) Gd/Yb vs La/Sm . Data sources: [1] Svensson (1998); [2] Liu et al. (1993), Gallet et al. (1996), Jahn et al. (2001), Ding et al. (2001) and Yokoo et al. (2004); [3] and [4] Honda et al. (2004). Compositional fields (ellipses) include data points and errors.

and 25%, respectively. A large mass of dust was therefore deposited during the 4-day transit. Since the path of the dust cloud crossed the Wrangell and St. Elias Mountains, part of the “missing” dust was most likely deposited there. How much of it was dry- or wet-deposited is uncertain, but orographic forcing of precipitation likely enhanced wet deposition through in-cloud or below-cloud scavenging.

Ground observations as well as model simulations of the April 2001 Asian dust outbreaks indicate that emissions originated from two broad regions: The Taklimakan and Tsarim Basin deserts of northwestern China, and the high-dust deserts of northern China and Mongolia, including the Gobi, Badain Jaran and Tengger deserts (Gong et al., 2003; Zhang et al., 2003b; Uno et al., 2004). However the climatology of the event was complex, and dusts from these two regions were not necessarily mixed and transported along the same pathways or over the same distances. Our own data from the St. Elias Mountains indicate that the part of the dust cloud

Fig. 8. (a) Sr–Nd and (b) Pb isotope ratios in dusts from the St. Elias Mountains (sites KC, MQM and QSB) compared with those in Yukon loess, Asian dust in Greenland ice [1, shaded], and potential Chinese desert sources [2–9]. All isotopic data used are from carbonate-free material, except for the dust sample from site KC (bulk). The bold dashed curve is an isotopic mixing line between a Chinese desert dust end-member ($^{87}\text{Sr}/^{86}\text{Sr} = 0.71511$; $\epsilon_{\text{Nd}_0} = -6.30$) and a Yukon loess-like end-member ($^{87}\text{Sr}/^{86}\text{Sr} = 0.70552$; $\epsilon_{\text{Nd}_0} = 1.47$). Data sources: [1] Biscaye et al. (1997), Svensson (1998) and Bory et al. (2003a); [2–9] Jones et al. (2000), Biscaye et al. (1997), Bory et al. (2003a) and Nakano et al. (2004).

which was transported across the subarctic North Pacific originated primarily from the Gobi desert region of northern China and Inner Mongolia. This is also consistent with previous trans-Pacific dust events (e.g. McKendry et al., 2001) and with climatological analyses which point to northern China and Inner Mongolia as the dominant sites for large-scale dust cloud generation (Wang et al., 2004b).

The April 2001 dustfall deposited large quantities of minerogenic elements over the central St. Elias Mountains, significantly altering the chemical composition of the snow cover. Our data show, for example, that [Pb] in the dust-impacted snow averaged 3 ng g^{-1} (ppb), which is ~ 10 times the largest annual mean [Pb] of 0.3 ppb measured by Hinkley (1993) in snows of the southeast Alaskan interior. Previous LRT events have shown that Asian dust clouds can be mixed with sulphate, elemental carbon (soot) or

trace metals from industrial sources (Nishikawa et al., 1991; Perry et al., 1999; Jaffe et al., 2003a). To check for the presence of pollutants in the snow impacted by the April 2001 dustfall, we calculated enrichment factors (EF) for 23 elements, including As, Cd, Cu, Ni, Pb, V and Zn. Because trace element data from eolian sediments are scarce, we normalized our data to UCC abundances (Taylor and McLennan, 1985) using Al as the reference element. Accordingly, for any element x in a sample, the enrichment relative to UCC is defined as $\text{EF}(x) = [x/\text{Al}]_{\text{sample}} \div [x/\text{Al}]_{\text{UCC}}$.

Results (Fig. 9) show that most elements in the particulate dust ($>45 \mu\text{m}$) deposited in the southern Yukon snows either have $\text{EF} \approx 1$, or are slightly enriched relative to UCC ($1 < \text{EF} \leq 10$), the most enriched being As, Cd and Sb. In contrast, Ca, Na, Sr and Pb are slightly depleted relative to UCC ($\text{EF} < 1$). The same pattern of enrichment (or depletion) was found if we used the total elemental concentrations measured in snow. Given that northern Chinese deserts soils contain abundant carbonates and evaporites, the low EF for Ca, Na and Sr in the Yukon snows suggest that some of these elements were lost from the dust during LRT, possibly by dissolution of carbonates and salts in precipitation (as proposed earlier). Losses during transport could also explain the low EF for Pb (≈ 0.3), or it may simply be that the Asian dust source is naturally Pb-depleted relative to UCC. For most other elements, the EF fall with-

Fig. 9. Enrichment factors relative to upper continental crust (EF_{UCC}) for selected elements in dusts sampled in the St. Elias Mountains (a and b), compared with (c) mineral aerosol data from central and northern China (Zhang et al., 1993, 2003b; Zhou et al., 1996; Mori et al., 2003). Vertical bars give the spread of data (min, max). The dashed line denotes $\text{EF}_{\text{UCC}} = 1$.

in or below the measured range in Chinese desert aerosols sampled at or near their sources (Zhang et al., 1993, 2003b; Zhou et al., 1996; Mori et al., 2003). Hence, we find no clear evidence of metal pollution transport in the dust-impacted snows of the St. Elias Mountains.

Asian dust storms can also deliver large inputs of iron to the Ocean, thereby enhancing the productivity of marine phytoplankton (e.g., Bishop et al., 2002). Following the passage of the April 2001 dust cloud, Johnson et al. (2003) reported a substantial increase in dissolved [Fe] in the northeast Pacific gyre, which they attributed to eolian inputs from the Asian dust cloud. They also commented that [Fe] in dust aerosols sampled at the Mauna Loa Observatory (MLO; Hawaii) during this period were too low to explain the observed Fe enhancement in Pacific gyre waters. However, [Fe] in MLO aerosols are measured in the $<2.5\ \mu\text{m}$ fraction (Perry et al., 1999). If the dusts deposited in the Pacific gyre had the same source and size distribution as those found on Mt. Logan (Fig. 3), the MLO aerosol data may considerably underestimate the eolian flux of Fe to the Ocean. This could account for part of the discrepancy noted by Johnson et al. (2003).

6. Summary and conclusions

In early May 2001, a widespread layer of mineral dust was discovered in the snows of the central St. Elias Mountains, southern Yukon Territory, Canada. A detailed analysis of the physico-chemical properties of these dusts confirm that they originated from the Gobi desert region of northern China and Inner Mongolia, and were transported across the North Pacific Ocean following an exceptionally intense series of dust storms that had taken place earlier in April. The most unequivocal Asian-source signatures were found in dusts deposited on the Mt. Logan massif at altitudes $\geq 3000\ \text{m}$. The dusts had a volumetric modal size of $\sim 4\ \mu\text{m}$, consistent with published data on far-traveled dusts found in alpine snowfields. However a sample taken at 5340 m had a much coarser size distribution that suggests rapid trans-Pacific transport of large mineral particles ($>10\ \mu\text{m}$) in the mid-troposphere immediately following the April 2001 dust storms.

The mass of dust that fell over the southwestern Yukon ($\sim 21,000\ \text{km}^2$) in April 2001 was estimated between 5500 and 6335 tons, the bulk of which may have been deposited over a ~ 1 -week period. Snowpack conditions and meteorological conditions at the time of the dustfall suggest that orographically enhanced snow scavenging was the dominant mechanism of dust deposition. Crustal enrichment factors of 20 elements (including toxic metals such as As, Cd and Pb) measured in the dust were within or below the range observed in Asian mineral aerosols. Hence, although the dust cloud was reportedly mixed with volatile pollutants from Asian (Jaffe et al., 2003; DeBell et al., 2004), our data provide no evidence that metal pollutants

were transported or deposited with the dust during this particular LRT event.

Recent climate warming, combined with unchecked land use and spreading desertification are being blamed for a recent rise in the frequency and magnitude of dust storms in East Asia, with major adverse consequences on the health and the economy of China and regions downwind from it, such as Korea (Chun et al., 2001; United Nations Environmental Programme, 2004). New ice-core records of atmospheric chemistry from the St. Elias icefields may soon help to verify if and how these changes are affecting the free troposphere across the northeast Pacific (Osterberg et al., 2005). It is hoped that data from our study may assist in detecting the signature of recent and ancient Asian dust LRT events in ice cores, thereby helping to develop a long-term proxy record of their frequency, magnitude and source(s).

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at [doi:10.1016/j.gca.2006.05.005](https://doi.org/10.1016/j.gca.2006.05.005).

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