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1 **Biomass-derived provenance dominates glacial surface organic**
2 **carbon in the western Himalaya**

3 SARWAR NIZAM^{1*}, INDRA S.SEN¹, VELU VINOJ², VALIER GALY³, DAVID SELBY^{4,5}, MOHAMMAD
4 F. AZAM⁶, SATYENDRA K. PANDEY², ROBERT A. CREASER⁷, AVINASH K. AGARWAL⁸, AKHILENDRA
5 P. SINGH⁸, MICHAEL BIZIMIS⁹

6 *¹Department of Earth Sciences, Indian Institute of Technology Kanpur, Kanpur, UP 208016,*
7 *India.*

8 *²School of Earth, Ocean and Climate Sciences, Indian Institute of Technology Bhubaneswar,*
9 *Odisha, 752050, India.*

10 *³Department of Marine Chemistry and Geochemistry, Woods Hole Oceanographic Institution*
11 *(WHOI), 266 Woods Hole Road, Woods Hole, MA 02543, USA.*

12 *⁴Department of Earth Sciences, University of Durham, Durham DH1 3LE, UK.*

13 *⁵State Key Laboratory of Geological Processes and Mineral Resources, School of Earth*
14 *Resources, China*

15 *⁶Discipline of Civil Engineering, School of Engineering, Indian Institute of Technology Indore,*
16 *Indore 453552, India.*

17 *⁷Department of Earth & Atmospheric Sciences University of Alberta, 126 ESB Edmonton Alberta*
18 *Canada T6G2R3.*

19 *⁸Engine Research Laboratory, Department of Mechanical Engineering, Indian Institute of*
20 *Technology Kanpur, UP, 208016, India.*

21 *⁹School of Earth, Ocean and Environment, University of South Carolina, Columbia, SC 29208,*
22 *USA.*

23 **Correspondence to: sarwar@iitk.ac.in*

24 Abstract

25 The origin, transport pathways, and spatial variability of total organic carbon (OC) on the western
26 Himalayan glaciers is poorly understood compared to that of black carbon (BC) and dust, but it
27 is critically important to evaluate the climatic role of OC in the region. Applying the distribution
28 of OC activation energy, ^{14}C activity and radiogenic isotopes of $^{208}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and
29 $^{206}\text{Pb}/^{204}\text{Pb}$ in glacial debris and atmospheric particulate matter (PM_{10} size fraction) we
30 demonstrate that $98.3 \pm 1.6\%$ and $1.7 \pm 1.6\%$ of the OC in western Himalayan glaciers are derived
31 from biomass and petrogenic sources, respectively. The $\delta^{13}\text{C}$ and N/C composition shows that
32 the biomass is a complex mixture of C3 vegetation and autochthonous photo-autotrophic inputs
33 modified by heterotrophic microbial activity. The dataset reveals that the studied western
34 Himalayan glacier has negligible contributions from fossil fuel derived particles, which contrasts
35 to the central and eastern Himalayan glaciers that have significant contributions from fossil fuel
36 sources. We show that this spatial variability of OC sources relates to regional differences in air-
37 mass transport pathways and precipitation regimes over the Himalaya. Moreover, our
38 observation suggests that biomass-derived carbon could be the only primary driver of carbon-
39 induced glacier melting in the western Himalaya.

41 1. Introduction

42 Worldwide glaciers are losing mass at an average rate of 0.48 ± 0.20 meter water-
43 equivalent per year (m w.e. yr^{-1})¹, with the rate of Himalayan glacier mass wastage nearly
44 doubling (-0.43 ± 0.14 m w.e. yr^{-1}) in recent years.² This accelerated mass loss is considered to
45 be primarily related to the well-established long-term increase of the Earth's near surface
46 temperature and anthropogenic climate change.^{3,4} In addition to this warming, the reduction of
47 surface ice albedo by ice surface deposits of dark colored impurities constitutes an additional
48 control of glacier melting rate.⁵⁻⁷ Impurities that darken the ice surface and directly increase the
49 heat absorption and thus enhance ice melting comprise dust, BC (black carbon, e.g., soot), and
50 other forms of OC not derived from combustion.⁸⁻¹¹ In this study, OC refers to the total organic
51 carbon that comprises the entire pool of organic carbon including both organic and elemental
52 carbon. Further, we classify the OC as OC_{ff} (fossil fuel) and OC_{bio} (biomass) referring to OC
53 derived from either fossil fuel or biomass sources, such as biomass burning derived particles,
54 atmospheric organic matters, and glacial microbes.⁵

55 The contribution to glacier surface darkening by particle matter and its attendant impact
56 on glacial melt across high Asian mountain glaciers is variable. For example, the main
57 contributor to glacier surface warming in Eastern Himalaya and Central Asia is BC, while
58 mineral dust is a dominant factor in the western Himalaya.¹² As a result, previous studies have
59 mainly focused on BC and dust, owing to their greater heat absorption capacities.¹³⁻¹⁶
60 Heretofore, limited research on total OC has led to substantial uncertainties and incomplete
61 knowledge of the impact of OC in the Himalaya and Tibetan Plateau.¹⁷⁻¹⁹ Thus, given the almost
62 20% albedo reduction related to OC for glaciers in Asia¹², a thorough assessment of the origin,
63 transport pathways, and spatial distribution of OC is paramount to reduce the uncertainties in

64 estimating the impact of carbon on the rate of glacial melting of Himalayan glaciers. In addition
65 to the albedo effect of light absorbing impurities, carbonaceous aerosols also warm up the air
66 mass over glaciers, and support microbial life²², both of which further contributes to enhanced
67 rates of glacier melting. It is also noteworthy that algal or microbial communities on the glacier
68 surface act as a sink for carbon and other impurities, and may yield a greater positive feedback
69 on glacier melting when compared to OC_{ff}.²³ As such, constraints on the composition of OC will
70 help better quantify the future impact of increasing anthropogenic emission surrounding
71 Himalayan glaciers (Figure S1), and its attendant controls on the hydrological regimes of glacier-
72 sourced large river systems such as the Indus, Ganges, and Brahmaputra. The findings will also
73 aid in quantifying the contribution of Himalayan glaciers melt to eustatic sea-level rise¹,
74 currently estimated to raise sea level by 0.52 cm by the end of 21st century.²⁴

75 In contrast to model-based approaches²⁶⁻²⁸, recent radiocarbon measurements on BC
76 particles deposited on glaciers and in aerosols revealed a much higher (~50%) fossil fuel
77 contribution across the Himalayan-Tibetan Plateau.²⁵ These radiocarbon measurements were
78 however restricted to the central and eastern Himalayan region, which falls under the strong
79 influence of the Indian Summer Monsoon. Further, the eastern and western areas of the
80 Himalayan region have different climatology, hydrology, meteorology, as well as glacier
81 behavior and dynamics.²⁹⁻³¹ The large geographical spread (~2500 km) of the Himalaya over
82 different climate regimes makes it very difficult to extend the findings of one climate regime to
83 the other. Moreover, in-situ OC measurements of glacier and snow samples in the western
84 Himalaya including Hindu-Kush and Karakoram region are not available, despite the fact that the
85 western region holds >70% of the total ice mass of the Himalaya.³² With one-sixth of the world's
86 population dependent on the Himalayan glacier meltwater and the increase in anthropogenic

87 emissions in the Indian subcontinent (Figure S1), understanding the origin and transport
88 pathways of OC on the western Himalayan glaciers and its impact on glacier melting rate is an
89 issue of global significance.

90 To understand the origin, transport pathways and spatial variability of OC on the western
91 Himalayan glaciers, this study focuses on the source area and origin of dust and OC on the
92 ablation zone (>4500 m a.s.l.) of the Chhota Shigri Glacier (CSG, 32.2° N, 77.5° E) in the
93 Lahaul-Spiti valley of the western Himalaya, India (Figure 1). Cryoconite and supra-glacial
94 moraine sediments from the CSG surface were investigated to characterize the origin of
95 carbonaceous particles. Cryoconites are small water filled depressions containing a dark colored
96 mixture of dust, OC, BC, and microbes collected over several years. Further, to constrain the
97 source end-members, atmospheric particulate matter (PM₁₀) were collected in Harsil (31.1° N;
98 78.7° E; 2634 m a.s.l.) and Kanpur (26.1° N; 80.2° E). The Harsil site is located in a high altitude
99 remote Himalayan forested area that records a history of major forest fires (Figure S3). Harsil is
100 >125 km from major urban settlements and industrial activities, thus the collected aerosol at
101 Harsil serves as the best proxy for the OC_{bio} and pristine Himalayan end-member. To obtain the
102 best estimate of OC_{ff} aerosol, a sample was collected from ~5 Km north of a coal-fired thermal
103 power plant (Panki Thermal Power Plant) and a kilometer west of a National Highway (NH91)
104 in Kanpur –one of the largest industrialized centers in the Indo-Gangetic plain (Figure 1).
105 Analysis of samples followed a multi-disciplinary approach, coupling organic and inorganic
106 geochemical tracers including Ramped Pyrolysis Oxidation (RPO), ¹⁴C activity, δ¹³C,
107 ²⁰⁸Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb, ²⁰⁶Pb/²⁰⁴Pb, TOC, N, and heavy metal concentrations with Hybrid Single
108 Particle Lagrangian Integrated Trajectory Model (HYSPPLIT) air mass trajectory modeling was
109 used to determine OC origin and transport pathways identification.

110 **2. Materials and Methods**

111 **2.1 Sampling details**

112 Cryoconite debris was collected from 20 sites in the ablation zone (4515 - 4928 m a.s.l.) of
113 the CSG during July, 2017. In addition to cryoconite, 5 moraine debris samples were also
114 collected. The samples were first dried on a hot plate at ~70 °C to remove the moisture content.
115 Moraine debris samples were sieved to get bulk (DB <3 mm) and particulate (<63 µm: DF)
116 fraction for geochemical analysis. Bulk cryoconite and moraine debris samples (DB and DF)
117 were ground to a homogeneous powder (20µm size) in an agate mil for further chemical analysis.
118 Atmospheric particulate matter (PM₁₀ size fraction) sampling in Kanpur and Harsil were carried
119 out using high-volume (1000 L/min) atmospheric aerosol samplers (Envirotech PM₁₀ sampler,
120 model APM 460 DXNL). In Kanpur and Harsil, PM₁₀ sampler was operated for 24 (on January
121 1, 2016) and 48 hours (April 12 to April 14, 2016), respectively. Detailed sampling protocol
122 description is outlined in Nizam and Sen, 2018.³³

123 **2.2 Ramped Pyrolysis Oxidation (RPO), ¹⁴C ages, bulk δ¹³C, N and C analysis**

124 The RPO analysis was carried out at the National Ocean Sciences Accelerator Mass
125 Spectrometer facility (NOSAMS, Woods Hole Oceanographic Institution). The RPO instrument
126 involves continuous sample heating, with the CO₂ evolved being trapped for dual C-isotope
127 analysis. Importantly, RPO analyzes 100% of the OC in the sample, irrespective of the nature of
128 the OC. Two cryoconite (C9 and C14), one moraine (DF6), and two aerosol samples were
129 selected for RPO analysis. The instrumental makeup and analytical protocol adopted for RPO
130 analysis has been described in detail in previous studies.^{34,35} Briefly, 30–70 mg powder sample
131 aliquots were loaded into a pre-combusted (at 850 °C for 5 hours) quartz reactor, heated at a 5 °C
132 min⁻¹ ramp rate in a two-stage oven. This incremental heating leads to the release of CO₂, which
133 was cryogenically purified, trapped and flame sealed into a glass tube containing nuggets of Ag

134 (~10 mg) and CuO (~100 mg). The CO₂ concentration in the carrier gas (in parts per million by
135 volume, ppm CO₂) is continuously measured at a resolution of 1 second by an infrared gas
136 analyzer. CO₂ collected at user-defined temperature intervals is referred to as RPO fractions.
137 Each RPO fraction collected is graphitized with the radiocarbon abundance determined via
138 Accelerated Mass Spectrometry at NOSAMS and reported as fraction modern (F_m). Meanwhile,
139 a 10% split of each RPO fraction was used for stable isotopes ($\delta^{13}C$) analysis using a dual-inlet
140 Isotope Ratio Mass Spectrometer (IRMS).

141 The $\delta^{13}C$ values are expressed in part per mill (‰) notation relative to Vienna Pee Dee
142 Belemnite (VPDB). The F_m and $\delta^{13}C$ ratios of the RPO fractions are reported in Table 1. In
143 addition, bulk $\delta^{13}C$, total organic carbon (OC), total nitrogen (N) were also measured in all of the
144 cryoconite and selected moraine samples using IRMS at Woods Hole Oceanographic Institution
145 (Table S1). The bulk $\delta^{13}C$ value calculated as the mass weighted average of all RPO fractions
146 showed good agreement with the measured bulk $\delta^{13}C$ value, with the exception of moraine
147 sample DF6. The mismatch appears to be driven by the high $\delta^{13}C$ value of the first RPO fraction
148 collected for this sample. We inferred this enriched stable isotope composition is related to some
149 technical error during IRMS analysis. The results for all of the RPO (and IRMS) analyses were
150 blank and kinetic isotope fractionation corrected.³⁶ Corrected data was used to calculate the
151 activation energy distribution using the rampedpyrox python package³⁷, which has been
152 described in detail in previous studies.³⁵ Radiocarbon age (in ¹⁴C yr BP) was calculated using
153 corrected F_m values and the Libby half-life as: $\text{age} = -8033 \ln(F_m)$.

154 **2.3 Lead isotope analysis**

155 Ten cryoconite and two moraine samples (DF) were selected for Pb isotopes analysis.
156 The Pb isotopic data was obtained from the Radiogenic Isotope Facility from the Department of

157 Earth and Atmospheric Sciences at the University of Alberta. Approximately 4-12 mg of sample
158 powder (depending on lead abundance), was dissolved in ultrapure HF/HNO₃ at 100°C for 2
159 days. Sample solutions were then evaporated under ULPA-filtered air and converted to chlorides
160 using 6N HCl, then bromides using 2N HBr. The Pb was purified by standard anion exchange
161 chromatography using HBr and HCl as eluents under ULPA-filtered conditions. The isotopic
162 composition of Pb then measured by Nu Plasma MC-ICPMS in static analysis mode. The
163 measured Pb isotope ratios were corrected for instrumental mass bias using the agreed value for
164 ²⁰³Tl/²⁰⁵Tl ratio measured simultaneously with each Pb analysis.³⁸ Overall reproducibility of any
165 Pb isotope measurement is based on >6 years of analyses of SRM981 Pb isotope standard. At the
166 first uncertainty level, the reproducibility of the SRM981 measured isotopic ratios are:
167 ²⁰⁶Pb/²⁰⁴Pb = 0.016%; ²⁰⁷Pb/²⁰⁴Pb = 0.018%; ²⁰⁸Pb/²⁰⁴Pb = 0.018%. The most widely accepted
168 values for this Pb isotope standard are those determined by double-spiked TIMS analysis in
169 previous study³⁹, which are: ²⁰⁶Pb/²⁰⁴Pb = 16.936; ²⁰⁷Pb/²⁰⁴Pb = 15.489; ²⁰⁸Pb/²⁰⁴Pb = 36.701.
170 The absolute values of Pb isotope ratios for SRM981 determined during the course of the
171 analyses report here are: ²⁰⁶Pb/²⁰⁴Pb = 16.936; ²⁰⁷Pb/²⁰⁴Pb = 15.488; ²⁰⁸Pb/²⁰⁴Pb = 36.690. The
172 Pb isotope ratios are reported in Table 2.

173 **2.4 Heavy metal analysis**

174 Heavy metal concentration analyses were performed in Indian Institute of Technology
175 Kanpur on a Quadrupole Inductively Coupled Plasma Mass Spectrometer (ThermoFisher
176 Scientific, Q-ICP-MS) system. Briefly, approximately 25 mg of sample powder was digested
177 using a mixture of HF (3 parts) and HNO₃ (1 part) for. Six procedural blanks, Reference Material
178 SBC-1 (Shale) from US Geological Survey (USGS) were also digested following the same
179 procedures. The final concentrations were blank corrected using the average procedural blank

180 concentrations and matrix effect was corrected by In normalization (Table S1). Average blank
181 corrections were less than 1% for most of the elements. The measured Sc, V, Cr, Ni, Cu, Zn and
182 Pb concentration of SBC-1 were 20 ± 1 , 205 ± 3 , 101 ± 2 , 81 ± 1 , 30 ± 0.2 , 191 ± 2 and 33 ± 1 (1 SD,
183 $n = 6$), respectively, which is in close agreement with the USGS certified values of 20 ± 0.2 , $220 \pm$
184 1.4 , 109 ± 1 , 83 ± 0.8 , 31 ± 0.6 , 186 ± 2 and 35 ± 0.3 , respectively.

185 **2.5 Air Mass Back Trajectory Analysis**

186 Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) modeling
187 was used to compute the air parcel back trajectories for every hour for five different sites that
188 includes Leh, ChhotaShigri, Thorong, Qiangyong, and Yulong sites for the year 2016 (Figure 1).
189 The archived meteorological analysis from National Centers for Environmental Prediction
190 (NCEP) Global Data Assimilation System (GDAS) model was used to estimate the trajectories.
191 Annual as well as seasonal trajectory analysis was carried out to elucidate the major source
192 regions of air mass reaching the Himalaya (Figure 1 and Figure S2). A total of 24×365 or 8760
193 trajectory pairs were simulated for each of the sites. A density map was created at a resolution of
194 $0.25^\circ \times 0.25^\circ$ resolution to obtain fraction of total trajectories passing through each of the grid
195 cell. This provided an indication of the percentage or probability of influence of each grid point
196 to the air mass reaching the receptor site. Two contour intervals of 0.1 and 0.5 that represented
197 10 and 50% trajectories passing through any grid was created for each site.

198 **3. Results and Discussion**

199 The distribution of OC activation energy (E_a), $\delta^{13}\text{C}$, and ^{14}C content (expressed as
200 fraction modern, F_m) for cryoconite, moraine sediments (particulate fraction) and PM_{10} were
201 compared to fingerprint the sources of OC (Figure 2). Activation energy is a proxy for OC
202 reactivity, and each OC pool has a distinct E_a distribution reflecting its overall bonding

203 environment (e.g. molecular composition, association with mineral surfaces).³⁵ Using E_a , $\delta^{13}\text{C}$
204 and ^{14}C ages, it is thus possible to fingerprint the sources of OC. For example, biomass-derived
205 OC have lower E_a ($<150\text{ kJ mol}^{-1}$), high F_m (~ 1.0), and modern ages, whereas fossil-derived OC
206 possess higher E_a ($>150\text{ kJ mol}^{-1}$) and $F_m = 0$.⁴⁰ Among the fossil sources, OC derived from
207 Himalayan petrogenic sources (OC_{petro}) has a much higher E_a ($\geq 200\text{ kJ mol}^{-1}$) than fossil fuel
208 derived OC ($\text{OC}_{\text{ff}} < 200\text{ kJ mol}^{-1}$).^{40,41}

209 The PM_{10} collected at Harsil and Kanpur has distinct E_a distributions and ^{14}C
210 compositions (Table 1, Figure 2). The E_a distributions of Harsil and Kanpur PM_{10} OC show a
211 predominant peak at ca. 135 and 165 kJ mol^{-1} , respectively. The Harsil PM_{10} OC is also
212 characterized by young ^{14}C ages (RPO F_m values >0.94 ; bulk $F_m = 0.99$). A predominant E_a
213 peak at ca. 135 kJ mol^{-1} and young ^{14}C ages of Harsil PM_{10} OC clearly corresponds to OC_{bio} .⁴⁰ It
214 is possible that some of the particles could contain soil organic carbon (SOC). However, SOC is
215 expected to be pre-aged, as observed in mineral soils globally and suspended sediments of
216 Himalayan sourced rivers.^{42,43} Further, dust contributions from arid areas (e.g., Thar Desert)
217 would similarly be expected to carry an old ^{14}C signature owing to conditions promoting organic
218 matter preservation. Our data is not consistent with SOC inputs as it shows a flat, near-modern
219 ^{14}C ages across the E_a spectrum. In contrast, Kanpur PM_{10} OC is characterized by lower F_m
220 values ($0.4 < F_m < 0.8$; bulk $F_m = 0.64$) and corresponding older ^{14}C ages. A predominant E_a peak
221 at ca. 165 kJ mol^{-1} and older ^{14}C ages of Kanpur PM_{10} OC reveal significant contributions from a
222 fossil fuel source. The lower E_a peak in Kanpur PM_{10} OC (ca. 135 kJ mol^{-1}) also points towards
223 some OC_{bio} contribution. A simple binary mixing calculation revealed that the Kanpur PM_{10} OC
224 is composed of ca. 36% OC_{ff} and 64% OC_{bio} . The PM_{10} RPO data further suggest that OC_{bio} and

225 OC_{ff} have overlapping Ea distributions, although OC_{ff} is more heavily concentrated at higher Ea
226 values.

227 OC in cryoconite and moraine sediment have similar Ea distributions with peak reactivity
228 between ca. 125 kJ mol⁻¹ and 150 kJ mol⁻¹. The low Ea and high Fm values, and modern ¹⁴C ages
229 support that OC in these RPO fractions is derived from recently fixed OC sources (OC_{bio}, Fm~1,
230 modern age) such as biomass burning, autochthonous photoautotrophic biomass and
231 heterotrophic microorganisms. Overall, the Ea and ¹⁴C signature of cryoconite and moraine
232 sediments are similar to Harsil PM₁₀, confirming an overwhelmingly dominant OC_{bio} input to
233 both cryoconite and moraine sediments. A small portion of cryoconite and moraine OC is
234 characterized by high Ea (>180 kJ mol⁻¹) and low Fm values (older ¹⁴C ages). The presence of
235 aged OC exclusively at high Ea values however precludes OC_{ff} as a source, because Kanpur
236 PM₁₀ RPO data show that OC_{ff} is distributed over the entire Ea spectrum. Instead, we argue that
237 the high Ea RPO fractions contain a mixture of biomass and petrogenic (rock-derived, OC_{petro})
238 OC. Indeed, OC_{petro} has been shown to be characterized by high Ea (i.e. >200 kJ mol⁻¹) and Fm
239 values equal to 0.⁴⁰ A mass balance approach assuming a binary mixing of ¹⁴C dead OC and
240 biomass-derived OC with a ¹⁴C age equal to that of Harsil PM₁₀, shows that 1.7± 1.6% of OC in
241 cryoconite and moraine sediment is derived from ¹⁴C-dead sources. This proportion translates
242 into a C concentration of 0.02± 0.02%. Such a low concentration of ¹⁴C-dead OC is consistent
243 with the expected OC_{petro} concentration in the high-grade crystalline rocks of the CSG catchment
244 and Himalayan rocks in general.⁴⁴ It is also lower than the average OC content of bulk moraine
245 samples (0.064± 0.020%), which, based on bulk geochemical characterization are dominated by
246 OC_{petro}. We conclude that cryoconite and moraine sediment have negligible OC_{ff} and contain a
247 mixture of OC_{bio} (98.3± 1.6%) and minor OC_{petro} inputs (1.7± 1.6%).

248 Additionally, we used stable carbon isotopes ($\delta^{13}\text{C}$) to further establish the source of
249 OC_{bio} in the cryoconite and moraine sediment. The majority of the cryoconite samples have a
250 distinct $\delta^{13}\text{C}$ composition with respect to surrounding moraine sediments, as well as to
251 cryoconite samples from Greenland and the Arctic.^{45,46} The $\delta^{13}\text{C}$ in cryoconite samples ranges
252 between -24.9 to -18.2‰ (average $-21.8 \pm 1.7\%$, $n=20$, 1s.d.), which is higher than for moraine
253 sediments (-22.5 to -26.7‰, average $-24.2 \pm 1.3\%$, $n=10$, 1s.d.). In general, carbonaceous aerosol
254 derived from burnt C3 type biomass sources in the Indian subcontinent should have a $\delta^{13}\text{C}$ value
255 of ca. -26‰⁴⁷, which can exhibit a maximum fractionation up to 0.5‰ due to burning emission
256 effect.⁴⁸ Burning induced ^{13}C fractionation of C3 type vegetation is therefore insufficient to
257 explain the observed $\delta^{13}\text{C}$ enrichment in cryoconite. Therefore, it can be postulated that
258 cryoconite draws some of its enriched $\delta^{13}\text{C}$ signature from additional sources or processes.

259 The $\delta^{13}\text{C}$ enrichment in cryoconite samples can be best explained by contributions from
260 photo-autotrophic and heterotrophic micro-organisms in supraglacial cryoconite that produces
261 enrichment of N and $\delta^{13}\text{C}$ values.^{22,46} Biomass humification processes could also produce
262 enriched $\delta^{13}\text{C}$ values, but the RPO- ^{14}C data are incompatible with significant SOC inputs. The
263 N/C vs $\delta^{13}\text{C}$ plot (Figure 3A) suggests that cryoconite samples are composed of a mixture of ^{13}C
264 depleted and N-poor organic matter that could correspond to a primary C3-derived OC, with ^{13}C
265 enriched and N-rich organic matter that reflects microbial communities growing in the snow/ice
266 environment. According to the RPO data, the ^{13}C -rich, N-rich component is also enriched in ^{14}C
267 (i.e. it has been fixed recently) and relatively labile, further suggesting it corresponds to
268 microbial biomass and/or algae. However, finer apportionment of the different sources of
269 biospheric OC in cryoconite samples would require compound-specific biomarker data. To
270 summarize, our data suggest that the OC in cryoconite and moraine sediments is a complex

271 mixture of C3 type biomass and autochthonous photo-autotrophic inputs modified by
272 heterotrophic microbial activity. An overall biomass source signature is also supported by heavy
273 metal ratios (Zn/Pb vs Cu/Pb; Figure 3B), assuming that the metal ratios retained the source
274 signature during transportation and transformation changes.

275 In addition to carbon systematics and heavy metal concentrations, $^{206}\text{Pb}/^{204}\text{Pb}$ and
276 $^{208}\text{Pb}/^{204}\text{Pb}$ were used as additional independent tracers to identify the residues of fossil fuel
277 signature on the CSG. The radiogenic Pb isotopes (Table 2), as well as heavy metal enrichment
278 (these are) factor (Figure S4) also support the absence of a fossil fuel signature in cryoconite and
279 moraine sediment from the CSG. Moreover, the $^{206}\text{Pb}/^{204}\text{Pb}$, $^{208}\text{Pb}/^{204}\text{Pb}$ overlap with that of the
280 Himalayan crust (Figure 4). The highly linear correlation between $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$
281 further implies that these elements are sourced from two dominant endmembers. Mixing
282 calculations using Pb isotope systematics show a negligible anthropogenic contribution (<1%,
283 Figure 4). The triple lead isotope data ($^{207}\text{Pb}/^{206}\text{Pb}$ versus $^{208}\text{Pb}/^{206}\text{Pb}$) also supports the absence
284 of anthropogenic pollutant contribution in the studied glacier site (Figure S5). Our results differ
285 from previous findings that the Himalayan glaciers in general receive significant long-range
286 transported dust from Africa, Middle East, and Thar desert, as well as from the Indo-Gangetic
287 plain, India.^{49,50}

288 The OC activation energy distribution, ^{14}C , $\delta^{13}\text{C}$, $^{208}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$,
289 TOC, N, and heavy metal concentrations, therefore, suggest that the studied western Himalayan
290 glaciers have negligible contribution from OC derived from fossil fuel combustion sources. The
291 difference between our findings and a previous study reporting ~50% fossil fuel sourced carbon
292 contribution in central and eastern Himalayan glaciers²⁵ can be explained by air mass transport
293 pathways and rainfall intensity. A HYSPLIT air mass trajectory modeling results reveal that the

294 eastern and central Himalayan glaciers that have received up to half of the BC from fossil fuel
295 sources draw a significant fraction of their air mass from the heavily polluted Indo-Gangetic
296 Plains (IGP) (Figure 1). In contrast, annual and seasonal (Figure 1 and Figure S2) air mass back
297 trajectory modeling at the study site shows that the western Himalayan regions receive limited
298 air-mass transport from the polluted IGP and less rainfall when compared to central and eastern
299 Himalaya, confirming the limited transport of ambient anthropogenic pollutants from the IGP.

300 The estimated glacial mass loss rates between 2000-2016 in western, central, and eastern
301 Himalaya are similar (-0.40, -0.35, -0.53 m w.e yr⁻¹, respectively).⁵¹ Glacial mass loss rate
302 reported for the CSG (-0.56 m w.e.yr⁻¹ between 2002 and 2014)⁵² is slightly higher than regional
303 averages. Since glacial mass loss is similar across the Himalaya and CSG is essentially free of
304 OC_{ff}, our observation suggests that OC_{bio} would be the primary driver of carbon-induced glacier
305 melting in the western Himalaya. Direct aerosol measurements studies over the western and
306 northwestern Himalaya hill stations also supports a predominant burnt carbon biomass
307 source.^{53,54} Given that it is well known that OC_{ff} particles are more readily transported over long-
308 distances compared to OC_{bio}⁵⁵, and thus, if OC_{bio} were transported from geographically distant
309 sources, OC_{ff} particles were expected to be similar or higher in concentration compared to OC_{bio}
310 in western Himalayan glaciers. The near absence of OC_{ff} and prevalence of local air mass
311 circulation therefore indicate that carbonaceous particles in western Himalaya are only derived
312 from locally burnt biomass sources. Thus, given that OC emissions are projected to be 21-28 Tg
313 by 2050⁵⁶, leading to new climate policies to curb global carbon emissions in urban centers,
314 considering the impact of local carbonaceous aerosols on glacier mass loss in the western
315 Himalaya is necessary to model the change in the glacial mass of western Himalayan glaciers.

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327

328 **Contributions**

329 I.S.S. conceived the study. S.N. and M.F.A did the field work. S.N. performed laboratory
330 measurements. S.N., I.S.S., V.V. and S.K.P. developed the air mass back trajectory models. S.N.,
331 I.S.S., V.G., V.V., A.K.A., A.K.S. and D.S. analysed the data. R.A.C. and M.B. contributed to
332 radiogenic isotope analysis and discussions. S.N. and I.S.S. wrote the paper with input from all
333 authors.

334 **Supplementary Materials:**

335 Figures S1-S5 and Table S1

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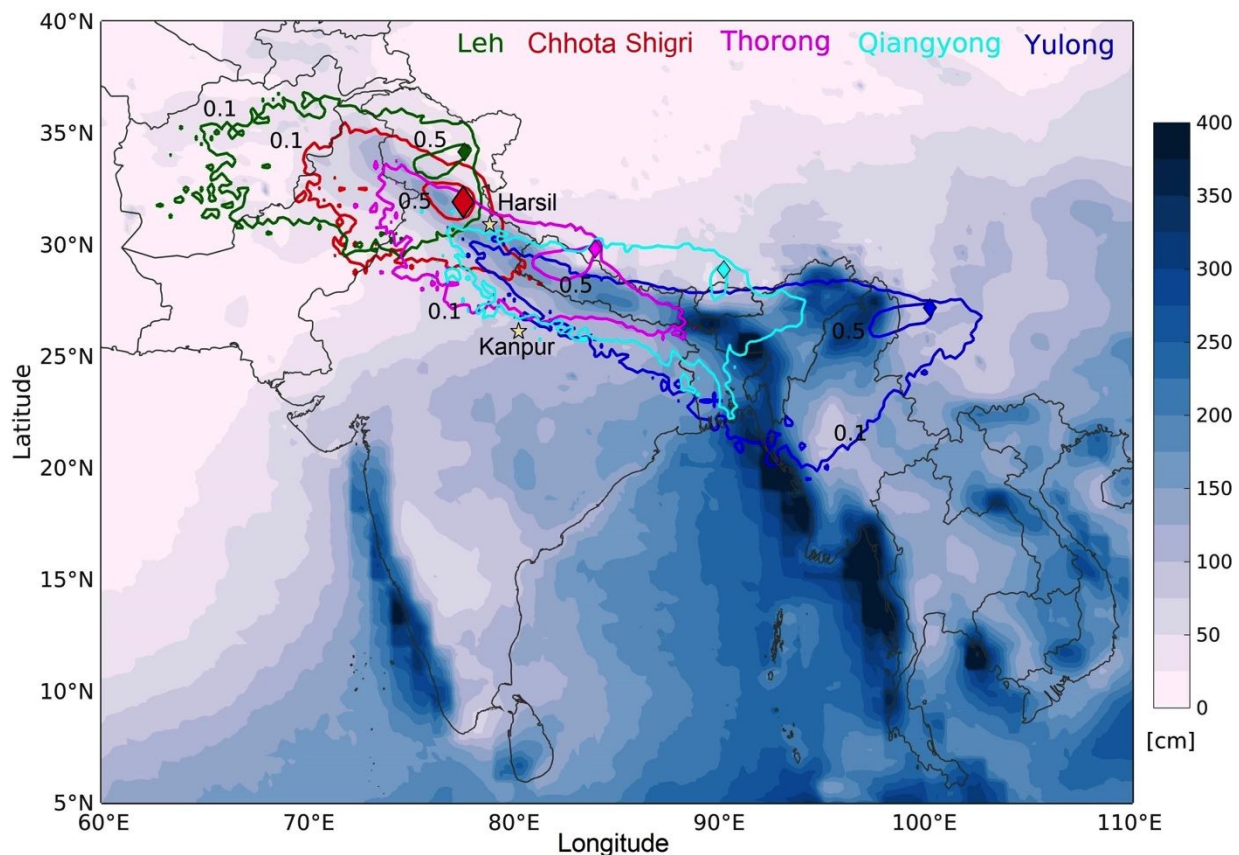
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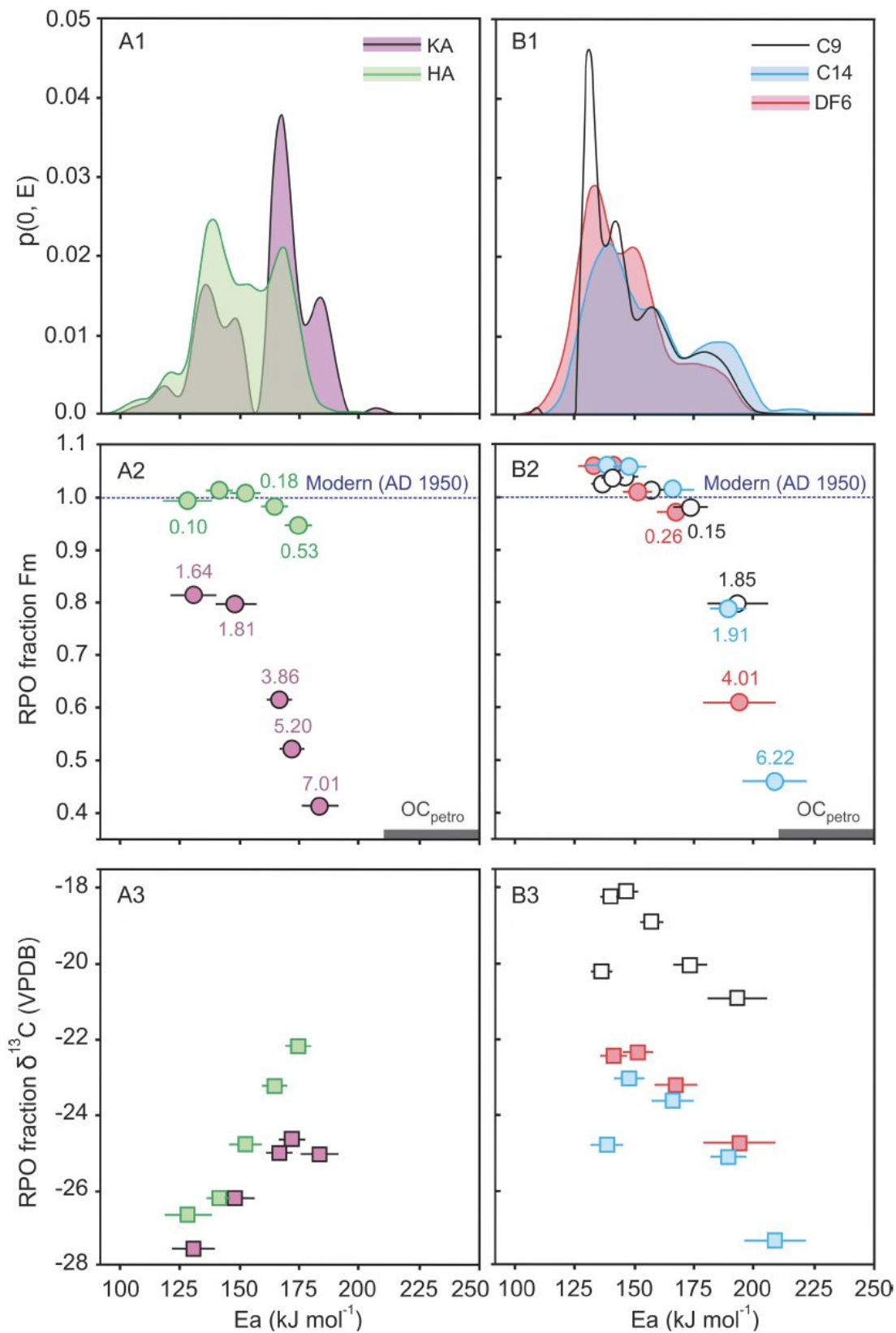
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635 **Figure 1. Climatological rainfall (TRMM_3B42_v7) with overlain trajectory density plot**
 636 **for few glaciers in the Himalayan-Tibetan Plateau region.** The line contours represent the
 637 fractional number of trajectories passing through a particular grid of spatial-resolution ($0.25^\circ \times$
 638 0.25°). For clarity, only two contours (of values 0.5 and 0.1) are shown for each receptor site.
 639 Here, 168-h HYSPLIT back trajectory starting from each of the five-receptor site for every hour
 640 (24×365) of the year 2016 were used. Thorong, Qiangyong, and Yulong sites have $\sim 50\%$ fossil
 641 fuel derived carbon.²⁵ Stars represent the aerosol sampling location and highlighted large
 642 polygon is our glacier-sampling site. The figure shows that there is a marked difference between
 643 the rainfall amounts between the western and eastern Himalayan glacier sites with eastern
 644 Himalaya obtaining higher rainfall. The 0.5 and 0.1 contour intervals show that the western
 645 Himalayan glacier sites are mostly influenced by air mass from western parts of the Himalaya,
 646 whereas central and eastern Himalayan glaciers are impacted by air mass from the Indo-Gangetic
 647 Plains (IGP) which is characterized by high level of anthropogenic activities (Figure
 648 S1). Seasonal trajectory density plots further show that wind trajectory origin is near similar
 649 irrespective of the seasons with most trajectories originating over a small region around the
 650 studied glacier (Figure S2).

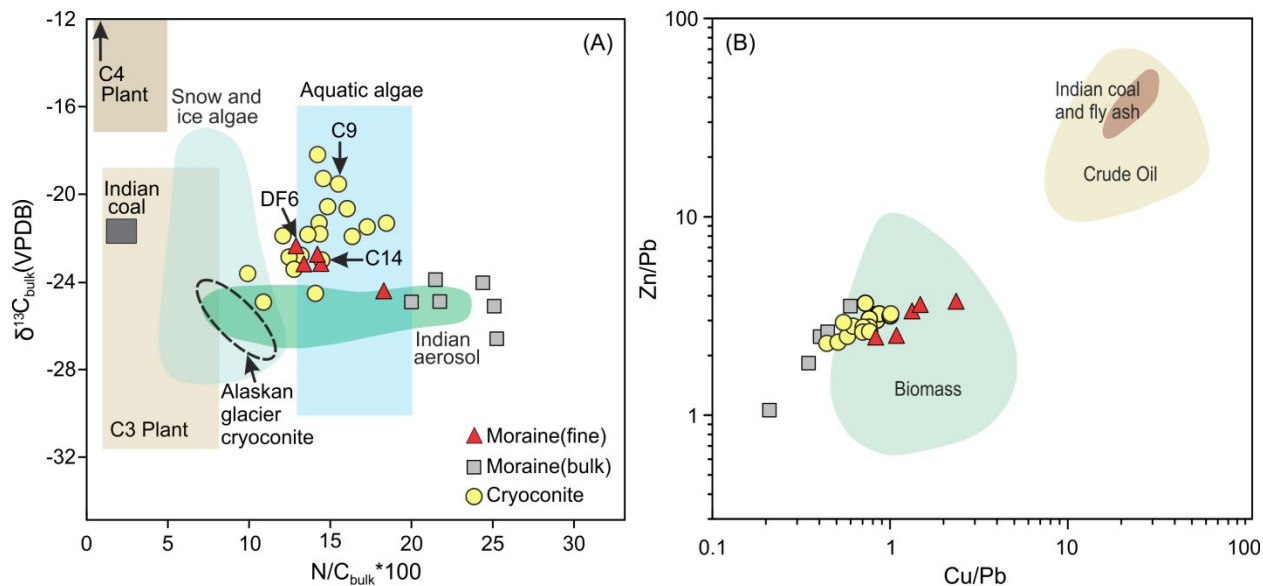


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653 **Figure 2. Apportionment of OC sources from RPO analysis.** OC activation energy (E_a)
654 distributions ($p(0, E)$) of (A1) aerosol (Kanpur Aerosol: KA, Harsil Aerosol: HA) and (B1)
655 glacier samples (cryoconite: C9 and C14, moraine fine fraction: DF6). Radiocarbon (Fm in
656 fraction) composition of each RPO fraction of the samples with associated radiocarbon age (in
657 cal kyr BP) and stable carbon ($\delta^{13}\text{C}$ in ‰) are plotted in middle (A2 and B2) and lower panel
658 (A3 and B3) respectively. Peak reactivity at ca. 165 kJ mol^{-1} and associated low Fm value and
659 old ^{14}C ages highlights OC_{ff} contributions in KA. Low OC thermal recalcitrance (peak $<150 \text{ kJ}$
660 mol^{-1}) and associated high Fm values and modern ages comparable to OC_{bio} and HA reflects
661 recently fixed sources. OC in glacier samples contain small contributions of high E_a material
662 reflecting binary mixing between a dilute petrogenic end member and a more concentrated
663 biomass/biospheric end member. Dashed line in both the middle panel represents modern
664 radiocarbon age.

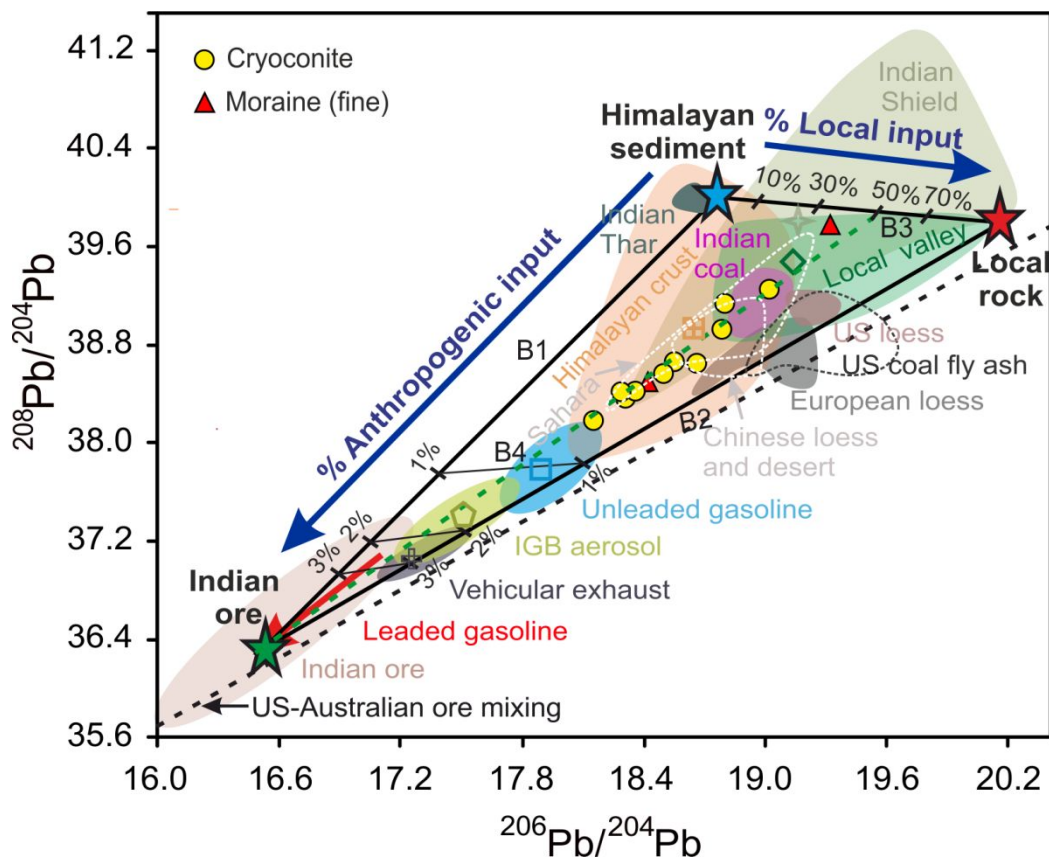
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668 **Figure 3. OC source inference from stable C isotopes, N/C and heavy metal ratios. (A)** Bulk
 669 OC, elemental (N/C) and isotopic ($\delta^{13}\text{C}$) mixing diagram showing the composition of cryoconite
 670 and moraine samples compared to that of tentative endmembers. Nitrogen and ^{13}C enrichment in
 671 cryoconite compared to bulk moraine suggests mixing between ^{13}C -poor, N-poor OM
 672 characteristic of biomass and ^{13}C -rich, N-rich OM typical of algae/microbes dwelling on glacier
 673 surface. Linear mixing trend between fine and bulk moraine suggests mixing between petrogenic
 674 and biospheric input. Literature references: C3 plant, C4 plant and aquatic algae⁵⁷, snow and ice
 675 algae^{22,58,59}, Indian aerosol.⁴⁷ **(B)** Heavy metal ratios in glacier samples compared to biomass,
 676 Indian coal, flyash⁶⁰ and crude oil.⁶¹



677
 678 **Figure 4. Mixing arrays in $^{206}\text{Pb}/^{204}\text{Pb}$ versus $^{208}\text{Pb}/^{204}\text{Pb}$ isotopic space.** The yellow circles and
 679 red triangles are new data points; literature data is shown with colored fields and open symbols
 680 within the field denotes their average composition. Thick black, dashed green and black lines are
 681 2-component mixing lines. The color-filled stars are end-member composition used in mixing
 682 calculations. The curves B1 and B2 defines mixing of Indian ore with Himalayan sediment (HS)
 683 and local crust (LC), respectively, while B3 represents mixing between LC and HS composition.
 684 Curves B4 is a mixing line between Indian ore with a sediment mixture comprising of 50% HS
 685 and 50% LC. We selected HS ($^{206}\text{Pb}/^{204}\text{Pb} = 18.81$, $^{208}\text{Pb}/^{204}\text{Pb} = 39.99$, Pb = 26.7 ppm)⁶² and
 686 LC ($^{206}\text{Pb}/^{204}\text{Pb} = 20.15$, $^{208}\text{Pb}/^{204}\text{Pb} = 39.79$, Pb = 33 ppm)⁶³ as a natural source end member.
 687 As an anthropogenic source end member representative, Indian ore ($^{206}\text{Pb}/^{204}\text{Pb} = 16.42$, $^{208}\text{Pb}/^{204}\text{Pb}$
 688 = 37.27, Pb = 4000 ppm)^{64,65} was chosen with the view that Pb used in various anthropogenic
 689 activities are mined from these ore inherit its source signature. Literature references for other
 690 regional source end-members are: Indian shield⁶⁶, Indian coal⁶⁷, US coal fly ash⁶⁸, IGB aerosol^{69–}
 691 ⁷², Sahara^{73,74}, European loess⁷⁵, Chinese loess and desert^{76–78}, Thar⁷⁶, US loess⁷⁸, unleaded
 692 gasoline⁷⁹, vehicular exhaust⁸⁰, leaded gasoline^{81,82} and US-Australian ore.⁸³

693 **Table 1.** RPO analysis results for cryoconite (C9 and C14), moraine (DF6) and aerosol samples
 694 (KA: Kanpur Aerosol, HA: Harsil Aerosol).

Sample-RPO fraction	T(°C)		m_F μg C	Ea (1 σ) (kJ mol ⁻¹)	Fm (2 σ) (fraction modern)	$\delta^{13}C$ (‰)	¹⁴ C age (calyr BP)
	min	max					
C9-F1	150	300	100.3	138 ± 6	1.0191 ± 0.0020	-20.57	>Modern
C9-F2	300	326	106.6	140 ± 4	1.0408 ± 0.0021	-18.37	>Modern
C9-F3	326	361	112.1	146 ± 7	1.0399 ± 0.0020	-18.07	>Modern
C9-F4	361	412	102.5	157 ± 8	1.0126 ± 0.0026	-18.77	>Modern
C9-F5	412	480	90.3	171 ± 9	0.9822 ± 0.0019	-20.03	145±15
C9-F6	480	794	88.0	191 ± 13	0.7947 ± 0.0021	-21.00	1850±20
C14-F1	150	318	136.4	136 ± 7	1.0563 ± 0.0021	-24.93	>Modern
C14-F2	318	375	135.0	148 ± 6	1.0529 ± 0.0021	-22.99	>Modern
C14-F3	375	465	135.0	165 ± 9	1.0137 ± 0.0023	-23.58	>Modern
C14-F4	465	560	100.0	188 ± 8	0.788 ± 0.0018	-25.1	1910±20
C14-F5	560	850	36.8	208 ± 14	0.4609 ± 0.0039	-27.17	6220±70
DF6-F1	150	298	108.0	132 ± 8	1.0461 ± 0.0022	-10.76	>Modern
DF6-F2	298	336	100.1	142 ± 6	1.0485 ± 0.0025	-22.43	>Modern
DF6-F3	336	387	109.0	153 ± 8	1.0063 ± 0.0023	-22.28	>Modern
DF6-F4	387	483	100.6	168 ± 10	0.9688 ± 0.0020	-23.17	255±15
DF6-F5	483	784	57.6	193 ± 15	0.6068 ± 0.0020	-24.71	4010±30
KA-F1	125	306	104.1	130 ± 11	0.8155 ± 0.0019	-27.66	1640±20
KA-F2	306	377	100.3	147 ± 10	0.7982 ± 0.0018	-26.17	1810±20
KA-F3	377	425	103.1	166 ± 6	0.6182 ± 0.0018	-25.11	3860±25
KA-F4	425	462	102.0	171 ± 6	0.5237 ± 0.0022	-24.60	5200±35
KA-F5	462	655	101.4	183 ± 8	0.4179 ± 0.0017	-24.91	7010±30
HA-F1	125	293	53.6	127 ± 11	0.9884 ± 0.0030	-26.83	95±25
HA-F2	293	335	51.9	142 ± 6	1.0096 ± 0.0036	-26.23	>Modern
HA-F3	335	385	52.8	152 ± 7	1.0029 ± 0.0033	-24.78	>Modern
HA-F4	385	439	52.1	165 ± 6	0.9775 ± 0.0033	-23.23	180±25
HA-F5	439	605	34.0	175 ± 7	0.9364 ± 0.0039	-22.13	530±35

m_F : Mass of carbon (as CO₂) contained in RPO fraction F

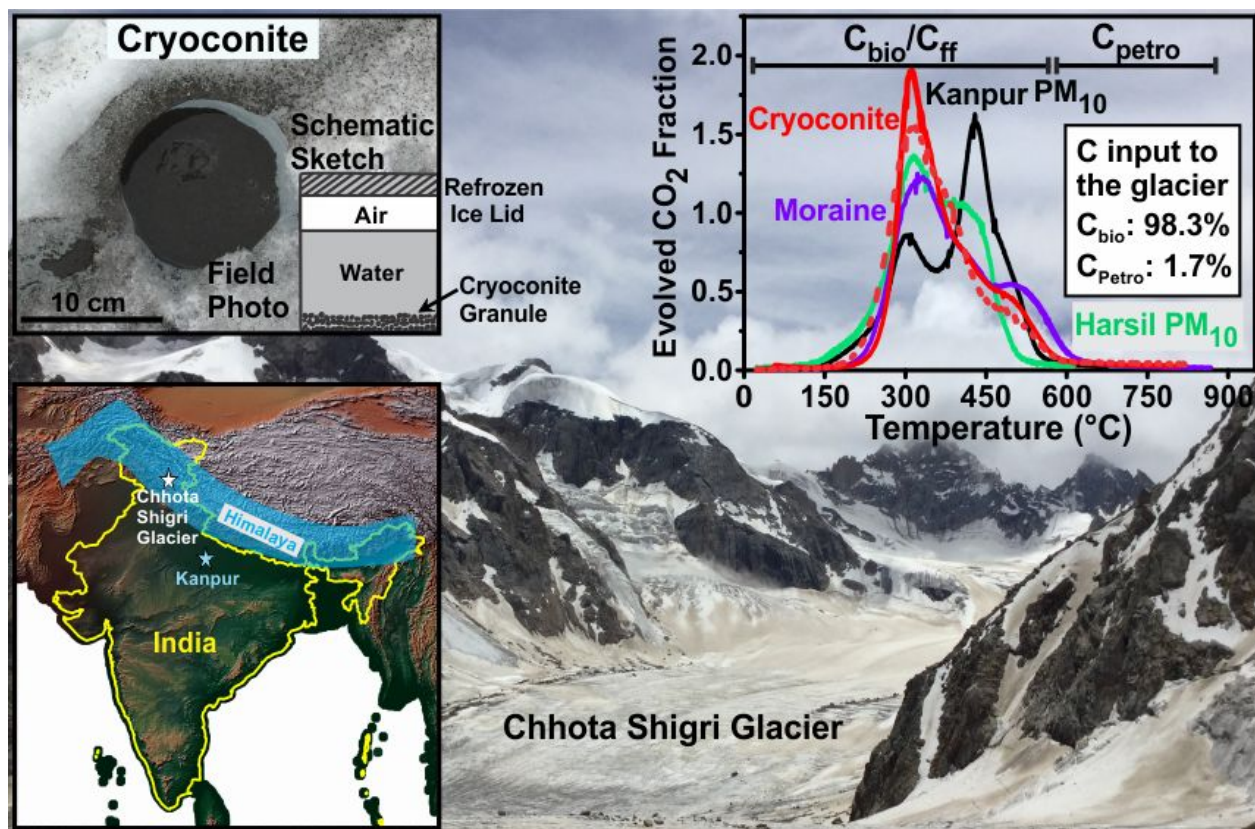
696 **Table 2.** Radiogenic lead isotope composition of selected cryoconite (Cx) and moraine (DF) samples.

Sample id	$^{208}\text{Pb}/^{204}\text{Pb}$	2SE	$^{207}\text{Pb}/^{204}\text{Pb}$	2SE	$^{206}\text{Pb}/^{204}\text{Pb}$	2SE	$^{207}\text{Pb}/^{206}\text{Pb}$	2SE
C1	38.91787	0.00546	15.70946	0.00182	18.78705	0.00159	0.83616	0.00003
C3	39.13549	0.00684	15.73477	0.00250	18.80094	0.00244	0.83691	0.00004
C5	39.23825	0.00680	15.74825	0.00218	19.02939	0.00224	0.82757	0.00005
C9	38.67497	0.00868	15.67102	0.00286	18.65789	0.00250	0.83991	0.00005
C11	38.67167	0.00520	15.68224	0.00166	18.55690	0.00178	0.84508	0.00003
C13	38.58314	0.00728	15.68314	0.00250	18.50767	0.00242	0.84740	0.00004
C14	38.38955	0.00552	15.65890	0.00204	18.32318	0.00202	0.85456	0.00003
C17	38.40762	0.00426	15.66683	0.00146	18.35741	0.00139	0.85343	0.00003
C18	38.40409	0.00630	15.65812	0.00242	18.30283	0.00202	0.85551	0.00004
C19	38.21132	0.00476	15.63569	0.00164	18.15303	0.00163	0.86133	0.00003
DF6	38.42784	0.00562	15.68993	0.00208	18.40597	0.00196	0.85239	0.00005
DF12	39.74669	0.00720	15.76498	0.00246	19.32824	0.00226	0.81567	0.00003

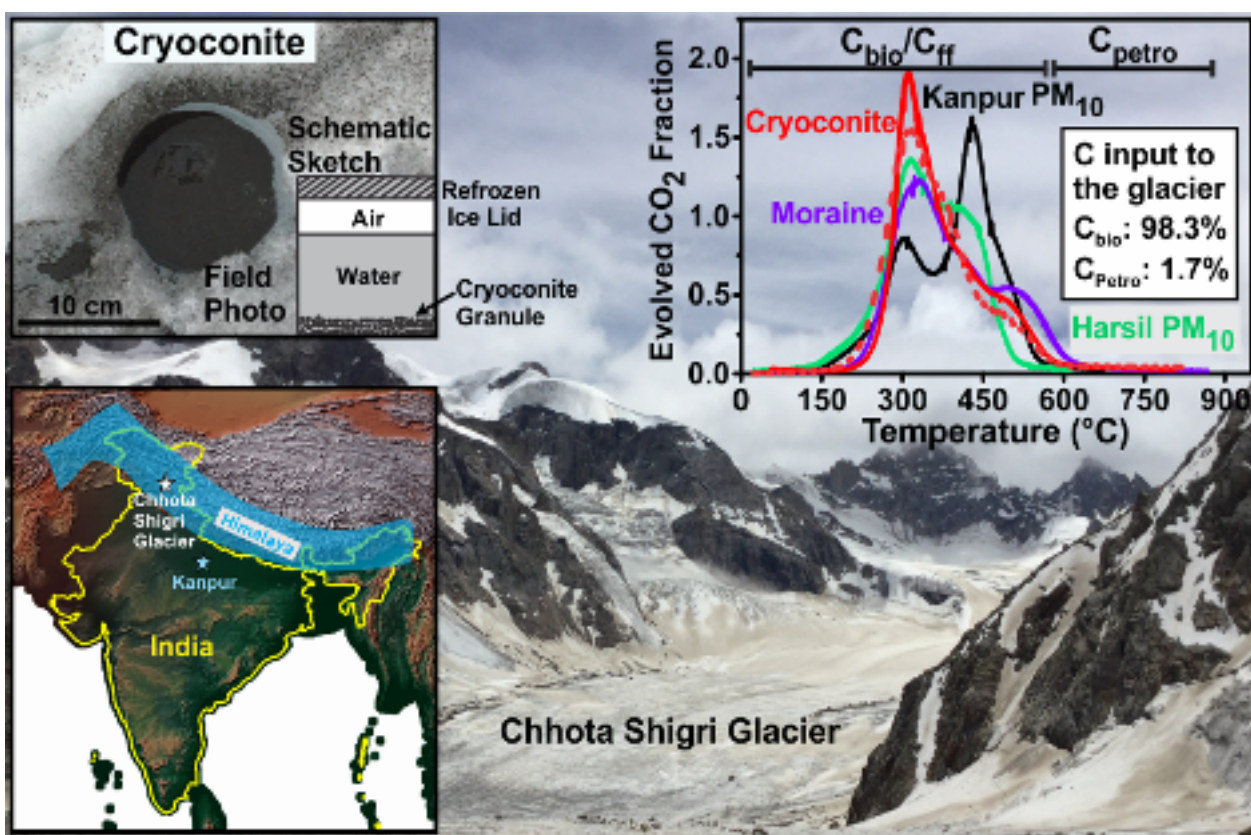
Note: Total Pb concentration in blank = 0.2 ± 0.16 ppb (1 S.D., n=4), Reproducibility uncertainty level = 1σ

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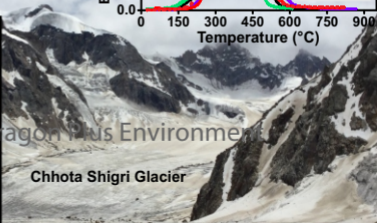
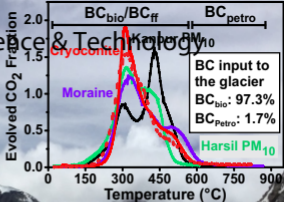
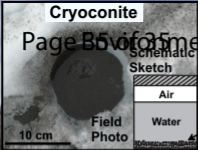


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Cryoconite

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