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The depositional flux of meteoric cosmogenic ¹⁰Be from modeling and

2	observation
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Abstract: Meteoric cosmogenic ¹⁰Be is a powerful tracer to quantify dates and rates of Earth surface processes over timescales of 10³-10⁵ yrs. A prerequisite for its applications is knowledge of the flux at which ¹⁰Be, produced in the atmosphere, is delivered to the Earth surface. Four entirely independent approaches are available to quantify this flux: 1) General Circulation Models (GCM) combined with ¹⁰Be production functions and aerosol dynamics; 2) ¹⁰Be in precipitation collections; 3) ¹⁰Be inventories in dated soil profiles; 4) riverine ¹⁰Be exported in solid and dissolved forms. We compiled and reprocessed published globally distributed ¹⁰Be flux data from each of these methods and compared them with each other after normalization to a common atmospheric production rate. Based on precipitation records, we propose a simple framework to discriminate between two delivery effects on ¹⁰Be fluxes. In the additive effect water vapor and ¹⁰Be are continuously accumulating during long-distance transport, leading to an increase in ¹⁰Be flux with precipitation rate. In the dilution effect, the ¹⁰Be flux is delivered from proximal vapor sources, limited by the rate of ¹⁰Be introduction from the stratosphere and independent of precipitation rate. Both effects are mostly present in combination, and the relative weight of either effect depends on vapor condensation rate and on the ratio of vapor condensation area to precipitation area. A comparison between precipitation-derived fluxes and GCM-derived fluxes shows that half of the precipitation estimates are >2 times greater than GCM-derived fluxes. By comparison, soil- and GCMderived fluxes agree within a factor of 2 for more than half (~57%) of the dataset, and the remaining soil estimates (~43%) are much lower than GCM-derived fluxes. 71% of ¹⁰Be flux estimates from riverine export using ¹⁰Be (meteoric)/⁹Be ratios also agree with GCM-derived fluxes within a factor of 2. We explain the precipitation-derived fluxes that commonly exceed all other estimates by short-term stochasticity in precipitation events that might introduce a measurement time-interval bias towards higher fluxes. This bias is not present over longer-term (10³-10⁵ yrs) flux estimates like those from soil profiles. Soil-derived fluxes might still present an underestimation when retention of ¹⁰Be in soil is incomplete. We recommend producing more ¹⁰Be depositional flux data from soil inventories with full Be retention, as these generate in our view the most relevant estimates for applications of meteoric ¹⁰Be on millennialscale Earth surface processes.

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- 40 **Keywords:** cosmogenic nuclides, meteoric ¹⁰Be, general circulation model (GCM), production rates,
- 41 Earth surface processes, geochronology

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1. Introduction

When galactic cosmic rays impinge upon the atmosphere, they induce spallation reactions in oxygen and nitrogen atoms that produce meteoric cosmogenic nuclide beryllium-10 (10Be) that is radioactive with a half-life of 1.39 Myr (Chmeleff et al., 2010). Meteoric ¹⁰Be is scavenged from the atmosphere primarily by precipitation after adhering to aerosols. When delivered to the Earth's surface it binds tightly to soil particles provided that the pH of soil solutions is circum-neutral. The concentration of meteoric ¹⁰Be measured in Earth surface materials is thus controlled by the following processes: ¹⁰Be production and redistribution in the atmosphere, ¹⁰Be delivery to the surface, ¹⁰Be retention in the substrate, and substrate stability with regards to erosion (Willenbring and von Blanckenburg, 2010b). As such, meteoric ¹⁰Be has been used as a tracer for a wide range of Earth surface processes over timescales of 10³-10⁵ yrs, including determining soil residence time (Bacon et al., 2012; Pavich et al., 1986) and ages of sediment archives (Egli et al., 2010; Lebatard et al., 2010), tracing soil movement (Jungers et al., 2009; Mckean et al., 1993), quantifying rates of erosion and weathering over hillslope scale (Maher and von Blanckenburg, 2016) and over basin scale (Brown et al., 1988; Portenga et al., 2019; von Blanckenburg et al., 2012; Wittmann et al., 2015), and also the expression of these processes back through time (von Blanckenburg et al., 2015; Willenbring and von Blanckenburg, 2010a). In order to quantify these dates and rates of Earth surface processes using meteoric ¹⁰Be, knowledge of the ¹⁰Be depositional flux is a key prerequisite.

¹⁰Be production in the atmosphere depends on the primary cosmic ray flux and thus on solar activity and geomagnetic field strength (Masarik and Beer, 2009). After production, ¹⁰Be is well-mixed in the stratosphere (~1 yr) and hydrolyzed to surface-reactive Be(OH)₂. After adsorption to atmospheric aerosols, ¹⁰Be is delivered to Earth's surface by wet and dry deposition. The delivery thus depends on

- atmospheric aerosol loading and climate, which can significantly modify the temporal and spatial distribution of ¹⁰Be depositional fluxes (Field et al., 2006; Heikkilä et al., 2013b). Over recent decades, substantial efforts on modeling and observation have been conducted to advance our knowledge of ¹⁰Be deposition over a range of temporal and spatial scales. These efforts include four independent approaches.
- 1) Atmospheric production and delivery models (Heikkilä et al., 2013a). In order to study production, atmospheric transport and deposition of meteoric ¹⁰Be, physics-based ¹⁰Be production functions (Masarik and Beer, 2009) and aerosol physics-chemistry (Stier et al., 2005) have been coupled with atmospheric general circulation models (GCM) (Schmidt et al., 2006; Stier et al., 2005). These integrated models have revealed the spatio-temporal pattern of ¹⁰Be depositional flux and its response to variations in solar activity, geomagnetic field strength (Field et al., 2006; Heikkilä et al., 2008b), and climate, including atmospheric mixing and precipitation (Heikkilä et al., 2013b; Heikkilä and Smith, 2013).
- 2) Precipitation collections. Over contemporary timescales, ¹⁰Be measurements in rainfall and snow in combination with precipitation rates yield local depositional fluxes. Based on such data, previous studies investigated the effects of latitude (Graly et al., 2011), altitude (Heikkilä et al., 2008a), stratosphere-troposphere exchange (Graham et al., 2003), precipitation rate (Willenbring and von Blanckenburg, 2010b), and aerosol content (Mann et al., 2011) on ¹⁰Be depositional fluxes.
- 3) Soil profiles. Over millennial timescales, inventories of meteoric ¹⁰Be in non-eroding soil profiles of known ages, developed on river terraces, moraine deposits, or bedrock, provide site-specific depositional fluxes that average over 10³ to 10⁵ years when assuming full retention (Dixon et al., 2018; Ouimet et al., 2015; Reusser et al., 2010). If ¹⁰Be retention is incomplete in soil profiles, this approach provides a lower-limit estimate (Maher and von Blanckenburg, 2016; Schoonejans et al., 2017).
- 4) Riverine fluxes. Over erosion and weathering timescales (e.g. 10³-10⁴ yrs), the atmospheric input flux of ¹⁰Be is expected to be balanced by the riverine sedimentary and dissolved ¹⁰Be flux (Wittmann et

al., 2015). Assuming such steady state, the ¹⁰Be flux from riverine export thus reflects the ¹⁰Be depositional flux over this long timescale.

Previous studies, including Willenbring and von Blanckenburg (2010b) and Graly et al. (2011), investigated the fluxes derived from different approaches by comparing modeled with measured ¹⁰Be depositional fluxes on the one hand, and short-term (precipitation-derived) with long-term (soil-derived) ¹⁰Be depositional fluxes in the other hand, respectively. In both types of comparison, the overall agreement between approaches was encouraging. However, in recent studies (Dixon et al., 2018; Ouimet et al., 2015), the ¹⁰Be depositional fluxes obtained from dated soil profiles clearly differed from flux estimates by GCM (Heikkilä and von Blanckenburg, 2015), and also from a fitting equation based on precipitation ¹⁰Be records (Graly et al., 2011). The lack of agreement between these estimates has been explained by local climatic variability (Heikkilä et al., 2013a), lack of suitability of soil inventories, or failure to adequately account for temporal variations in cosmic ray fluxes.

An understanding of the climatic control on ¹⁰Be depositional flux, meaning the relationship between ¹⁰Be concentration, flux, and precipitation rate, is essential for upscaling contemporary ¹⁰Be fluxes in space and time. In this regard, two contrasting views exist. One observation holds that the ¹⁰Be depositional flux is positively correlated with precipitation rate, and that this precipitation dependence can explain the variations of ¹⁰Be inventories in some soil profiles (Dixon et al., 2018; Graly et al., 2011). Other studies show no correlation between ¹⁰Be flux and precipitation, and regard the ¹⁰Be flux to be independent of precipitation (Ouimet et al., 2015; Willenbring and von Blanckenburg, 2010b). To account for these disparate views, Willenbring and von Blanckenburg (2010b) proposed two ¹⁰Be delivery effects: the "dilution effect" and the "additive effect". In the dilution effect, precipitation is delivered from moist air mass with proximal vapor sources where moisture has high turnover rates, such that the delivery of meteoric ¹⁰Be is ultimately limited by the rate at which it is introduced into moist air mass by stratospheric production. Hence the ¹⁰Be concentration in precipitation can be "diluted" (Willenbring and von Blanckenburg, 2010b). In the additive effect, water vapor is continuously accumulating in moist air

mass during long-distance transport, and so is ¹⁰Be, such that the ¹⁰Be depositional flux will increase with precipitation rate (Graly et al., 2011). However, the competition between both effects is still unclear and requires a comprehensive comparison between flux estimates across spatial and temporal scales.

In this study, we compiled literature data and recalculated ¹⁰Be fluxes recorded in different kinds of materials including precipitation, soil profiles and riverine solid and solute loads, and compared them to modeled (GCM-derived) ¹⁰Be depositional fluxes given the wide spatial coverage (i.e. global scale) of the GCM-derived dataset. The main aim is to evaluate the reliability of ¹⁰Be depositional fluxes from each approach and the associated bias in flux derivation. This study advances the understanding of meteoric ¹⁰Be delivery and deposition over a range of spatial and temporal scales, and provides new insights on method-specific uncertainties on ¹⁰Be flux estimates which shall benefit future Earth surface applications using meteoric ¹⁰Be.

2. Methods: compilation of modeled and measured ¹⁰Be fluxes and assessment of delivery effects

2.1. GCMs with ¹⁰Be production functions and aerosol dynamics

To investigate the spatio-temporal pattern of ¹⁰Be deposition, two GCMs, including GISS (Goddard Institute for Space Studies) ModelE (Field et al., 2006) and ECHAM5 (European Centre for Medium-Range Weather Forecasts-Hamburg Model 5)-HAM (Heikkilä et al., 2013a), were previously applied by incorporating meteoric ¹⁰Be production functions in the atmosphere (Masarik and Beer, 2009) and aerosol physics-chemistry. Comparing both models, the GISS ModelE with 20 vertical layers and a horizontal resolution of 4°×5° (Field et al., 2006) shows a coarser spatial resolution than the ECHAM5-HAM that includes 31-39 vertical layers and a horizontal resolution of 2.8°×2.8° (Heikkilä et al., 2008b). It has been suggested (Heikkilä et al., 2013a) that the lower spatial resolution of GISS ModelE (Field et al., 2006) might bias the modeling of atmospheric mixing and advection of ¹⁰Be, leading to artificial latitudinal differences in the response of ¹⁰Be deposition to production changes (Heikkilä et al., 2008b). We thus focus on the modeled ¹⁰Be depositional flux derived from the ECHAM5-HAM (Fig. 1).

We used published results of the ECHAM5-HAM model based on a ¹⁰Be production rate calculated for a modern solar modulation factor (φ in MeV) of 501.76 MeV (Fig. 2). To account for the climate-related uncertainty introduced by spatial variations in ¹⁰Be delivery over the Holocene, we used two different model outputs: one for modern ("industrial", time span: 1977-2006) climatic conditions (Heikkilä and Smith, 2013) and the other for the early Holocene ("pre-industrial", time span: 12-10 kyr BP) conditions (Heikkilä et al., 2013b). The difference in the spatial pattern of modeled ¹⁰Be depositional fluxes between both runs is our estimate of flux uncertainty (Heikkilä and von Blanckenburg, 2015). Here, the modeled fluxes of both runs under modern production conditions were rescaled to the average Holocene ¹⁰Be production rate (Fig. 2). The resulting distribution map (Fig. 1) shows the average flux of both rescaled model runs.

2.2. Precipitation collections

We compiled 10 Be data from precipitation collectors ([10 Be] $_{rain}$, in at/m 3) and the corresponding precipitation rate (P, in m 3 /m 2 /yr)) to calculate the 10 Be depositional flux (F $_{met}$, in at/m 2 /yr)):

$$F_{\text{met}} = \begin{bmatrix} {}^{10}\text{Be} \end{bmatrix}_{\text{rain}} \times P \tag{1}$$

The analytical uncertainty of [10 Be] rain was propagated into the uncertainty of F_{met} . To obtain an annual 10 Be depositional flux from measured monthly data, we calculated the monthly-precipitation-weighted 10 Be concentration and multiplied it by the annual precipitation rate. The derived flux is the sum of three components: wet deposition (scavenging of 10 Be(OH) $_2$ by precipitation), dry deposition (delivery by friction-based aerosol deposition), and the recycled fraction (10 Be carried by dust). This precipitation dataset covers continental settings (Fig. 1), including Switzerland (Heikkilä et al., 2008a), India (Somayajulu et al., 1984), and USA (Monaghan et al., 1986), and islandic settings, including Trinidad (Brown et al., 1992), Kikai Island (Japan) (Maejima et al., 2005) and New Zealand (Graham et al., 2003). Records with an observation period <9 months (see details in Supplementary dataset) are considered unrepresentative over the annual scale as they might be biased by seasonal cycles (Heikkilä and Smith,

2013), and thus these ¹⁰Be data are shown in figures for completeness but excluded from the discussion of fluxes.

We applied normalization procedures that account for variations in the flux of galactic cosmic rays and analytical differences, to compare between datasets of different observation periods and derived from different methods (Fig. 2). 1) All ¹⁰Be concentrations were normalized to the AMS (accelerator mass spectrometry) standard KNSTD07 (Nishiizumi et al., 2007). 2) Precipitation records that are subject to inter-annual variations in solar modulation were normalized to a common modern solar modulation factor of 501.76 MeV. 3) The contemporary precipitation-derived fluxes were further normalized to the average Holocene ¹⁰Be production rate by multiplying them with ~1.23 which is the ratio of Holocene relative to modern production and encompasses variations in solar modulation and magnetic field strength (Steinhilber et al., 2012) (Fig. 2).

2.3. Soil profiles

Over a timescale integrated by most geomorphic processes (10^3 - 10^5 yrs), the site-specific 10 Be depositional flux can be calculated from the total 10 Be inventory (at/m²) measured in soil profiles. Prerequisites are a known age of the onset of soil formation (t, in yr) and full retention of meteoric 10 Be. In the case of an alluvial substrate any meteoric 10 Be ([10 Be] $_{inh}$, in at/kg) inherited prior to deposition that is apparent at the bottom of profiles should be subtracted. The equation for calculating F_{met} as modified from Egli et al. (2010) is:

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$$F_{met} = \sum_{i=1}^{n} \left[\left(\begin{bmatrix} {}^{10}\text{Be} \end{bmatrix}_{s,i} - \begin{bmatrix} {}^{10}\text{Be} \end{bmatrix}_{inh,i} \right) \times \rho_i \times z_i \times V_{f,i} \right] \times \lambda/(1 - e^{-\lambda t})$$
 (2)

where [10 Be]_{s,i} is the soil 10 Be concentration (at/kg) at ith horizon, ρ_i the soil bulk density (kg/m³), z_i the thickness of the ith horizon (m), $V_{f,i}$ the volume percentage (%) of the fine earth fraction (<2 mm) containing 10 Be at the ith horizon, λ the decay constant of 10 Be (5×10⁻⁷ yr⁻¹). If no V_f data was reported in literature because the fine fraction dominates, V_f of 100% is used. The uncertainty of the resulting F_{met} is propagated from the individual uncertainties of the 10 Be measurement, the age determination, 10% for soil

density measurements, and 5% for V_f (Egli et al., 2010). We compiled 10 Be data from soil profiles (Fig. 1) located in continental settings, including European Alps (Egli et al., 2010) and the USA (Balco, 2004; Ouimet et al., 2015), and in islandic settings, including Hawaii (Dixon et al., 2018) and New Zealand (Reusser et al., 2010). We did not compile or show data from other archives like coral reefs (Maejima et al., 2005) or old soil profiles (age >200 ka) (Pavich et al., 1986) here because of 10 Be contamination by seawater and a high potential for surface erosion and incomplete retention, respectively. We applied 1) the AMS normalization same as above and 2) a millennial-scale variability normalization, depending on the integration timescale of each soil profile, to the average Holocene 10 Be production rate for comparison with other approaches (Fig. 2).

Some of the compiled profiles are considered to be affected by significant erosional loss, sampling depth not sufficient to quantify the ¹⁰Be inheritance, or excessive ¹⁰Be inheritance. These are shown in figures for completeness but excluded from the discussion of fluxes (see details in Supplementary dataset).

2.4. Riverine export

Over the river basin scale, when assuming steady state of ¹⁰Be fluxes over weathering and erosion timescales, the basin-averaged ¹⁰Be depositional flux can be derived from the ¹⁰Be flux exported by rivers (Brown et al., 1988). However, a major obstacle arises from the grain size dependence of ¹⁰Be concentrations in river sediment as Be adsorption depends on particle surface area (Singleton et al., 2016; Wittmann et al., 2012), more specifically, on adsorption site density (Maher and von Blanckenburg, 2016). As such, we adopted the isotopic ratio of ¹⁰Be to ⁹Be which is independent of mineralogical sorting, sorption capacity, and Be retentivity (Wittmann et al., 2012). We rearranged the equation for calculating denudation rates using ¹⁰Be (meteoric)/⁹Be ratios (von Blanckenburg et al., 2012) to solve for F_{met}:

$$F_{\text{met}} = D \times \left(\frac{^{10}\text{Be}}{^{9}\text{Be}}\right)_{\text{reac}} \times \left[^{9}\text{Be}\right]_{\text{parent}} \times f_{\text{reac+diss}}^{9} \times (1 - e^{-\lambda t_{s}})$$
(3)

where D (kg/m²/yr) is the independently known catchment-wide denudation rate, (10 Be/ 9 Be)_{reac} is the concentration ratio of 10 Be (at/kg) to 9 Be (mg/kg) measured in the reactive fraction of river sediments

(adsorbed onto or precipitated in secondary minerals). [9 Be]_{parent} (mg/kg) comprises the 9 Be present in the parent bedrock prior to weathering, determined by assuming a value of the average upper continental crust (2.5 ± 0.5 mg/kg) (von Blanckenburg et al., 2012) or by measuring local bedrock samples in small catchments (Dannhaus et al., 2018; Deng et al., 2020). f^{9} _{reac+diss} represents the mobile flux fraction of 9 Be that is released from primary minerals during weathering and measured in the sediment reactive fraction and dissolved fraction. Radioactive decay of 10 Be is negligible if the timescale of sediment transfer and storage (t_s) is short compared to its half-life.

We compiled literature meteoric ¹⁰Be and ⁹Be data of river sediment with *in situ* cosmogenic ¹⁰Be data nearby, which was used as an independent estimate of D in Eq. (3). This dataset covers a wide spatial range (Fig. 1), including the Amazon River basin in South America (Wittmann et al., 2015), the Potomac River basin in USA (Portenga et al., 2019), the Zhuoshui River in Taiwan Island (Deng et al., 2020), the Ganga River basin in South Asia (Rahaman et al., 2017), and small catchments in the Slavkov Forest of Czech Republic (Dannhaus et al., 2018). The reported water pH was >7, suggesting a low dissolved Be flux, in most of studied rivers except for catchments in the Czech Republic, where the dissolved Be loss was corrected to calculate f⁹_{reac+diss} (Dannhaus et al., 2018). All these ¹⁰Be measurements were based on the standard KNSTD07 or equivalents, and the used rock density was normalized to 2.65 g/cm³.

2.5 Discrimination of ¹⁰Be delivery effects

We discriminate between additive and dilution effects using a simple mass balance framework of water vapor and ¹⁰Be in the "source" (condensation) and the "sink" (precipitation). Details of mass balance equations are provided in Appendix 1. In general, the ¹⁰Be concentration of wet deposition is described by the ratio of the ¹⁰Be production flux from the upper atmosphere (F_{prod}, in at/m²/yr) to the condensation rate in the vapor source area (C, in m³/m²/yr) (Fig. 3). The difference in F_{met} between additive and dilution effects lies in the travel distance of moist air mass, illustrated by the ratio of surface area of condensation (A_c, in m²) to area exposed to precipitation (A_p, in m²). A_c/A_p ratios close to unity and >>1 indicate dominance of dilution effect and additive effect, respectively.

In a diagram of [10 Be]_{wet} versus 1/P, a dilution effect is illustrated by an A_c/A_p of 1 and co-variation of P with C, resulting in a slope (at/m²/yr) equal to the 10 Be production flux F_{prod}, i.e. a constant F_{met} (Fig. 3a) (Willenbring and von Blanckenburg, 2010b). In contrast, a pure additive effect results in a zero-slope relationship in that diagram where differences in P result from differences in A_c/A_p, and the y-intercept (at/m³) is equal to F_{prod}/C, resulting in a constant [10 Be]_{wet} for a given C (Fig. 3c).

In practice, the variable behaviors of moist air masses carrying both vapor and 10 Be will lead to a combination of delivery effects. When both A_c/A_p and C vary over the observation timescale, a mixture of dilution and additive effects may result. In this case, the 10 Be flux can be separated into two components: one component with unity A_c/A_p ratio (stable vapor source) and constant 10 Be flux (dilution component), and the other with constant condensation rate and thus constant $[^{10}$ Be]_{wet} (additive component). In the $[^{10}$ Be]_{wet} versus 1/P diagram the dilution component is identified from the slope (Fig. 3) and the additive component from the y-intercept. For a mixture of dilution and additive components, the proportion of the additive component (f_{add} , %) relative to the total 10 Be flux can be calculated as:

$$f_{add} = (intercept \times P)/(intercept \times P + slope) \times 100\%$$
(4)

This relationship shows that when P gets very high and there is a y-intercept resulting from the additive effect, intercept \times P >> slope and f_{add} will be close to 100%. The framework developed also allows taking dry and dust 10 Be components into consideration. In their presence the slope in the $[^{10}$ Be]_{wet-1}P diagram (Figs. 3a-c) will steepen, because dry-dust 10 Be depositional fluxes are independent of precipitation rate and contribute as a component diluted by precipitation. Although mixing between additive and dilution effects can be shown by modern precipitation records (Eq. (4)), this mixing process also presents in millennial-scale records and may vary with time.

3. Results: latitudinal distribution of ¹⁰Be depositional fluxes

All fluxes compiled from different approaches are shown as normalized to a common Holocene production rate in Fig. 4, and both the original and the re-normalized ¹⁰Be fluxes are provided in Supplementary dataset.

From the GCM output, the Holocene depositional fluxes, averaged over a given latitude band, are within a relatively narrow range for latitudes from 0° to 50°, ranging from 0.88×10^6 to 1.55×10^6 at/cm²/yr. The peak of GCM-derived flux is located at ~40° and can be explained by a higher air mass contribution from the stratosphere, which is the major reservoir (~65% of the total) of meteoric ¹⁰Be (Heikkilä et al., 2013a).

From precipitation collections we recalculated ¹⁰Be depositional fluxes based on Eq. (1). Contemporary ¹⁰Be depositional fluxes from reliable records (filled symbols) generally increase from low to high latitudes, with a range of 0.42 to 5.30×10^6 at/cm²/yr between latitudes of 10.4° and 47.3° . The reported percentage of recycled ¹⁰Be from eolian dust, calculated by different methods for the dataset in New Zealand (35-50°N), Switzerland (46-47°N) and USA (30-50°N), varies from 6.2% to 35% (see Table S1 in Supplementary materials). Given the differences in dust correction methods employed and the substantial uncertainties resulting thereof, total ¹⁰Be depositional fluxes are reported instead of fluxes corrected for recycled fractions. As such, these estimates can be directly compared to fluxes from soil profiles and riverine export where the recycled fraction is also included.

We also recalculated the ¹⁰Be flux for comparison based on an empirical equation for estimating primary ¹⁰Be fallout derived from data fitting between precipitation rate and precipitation ¹⁰Be fluxes at mid-low latitudes (Graly et al., 2011):

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$$F_{\text{met}} = P \times (1.44/(1 + EXP((30.7 - L)/4.36)) + 0.63) \times 10^{10}$$
 (5)

where L is latitude (°) and P is precipitation rate (m³/m²/yr). The recycled ¹⁰Be component (dust contribution) was subtracted here. The derived fluxes, originally rescaled to global production rate based on a φ of 700 MeV (Graly et al., 2011), were re-normalized to an average Holocene rate following

normalization steps 2) and 3) in Section 2.2 for comparison with other approaches. The latitudinal trend of precipitation collections is mirrored by this fitting equation (Graly et al., 2011) as expected (correlation: R = 0.60, p < 0.001) given that this fitting equation is derived from the same dataset.

Based on Eq. (2), we recalculated ¹⁰Be depositional fluxes from soil profiles. These millennial-scale fluxes from reliable records (filled symbols) vary from 0.12×10^6 to 4.07×10^6 at/cm²/yr. No clear trend with latitude is apparent from soil-derived fluxes as observations exist in only few latitudinal bands. Low soil pH (<4) may lead to non-negligible loss of dissolved ¹⁰Be (Maher and von Blanckenburg, 2016) and thus may bias the soil-derived ¹⁰Be flux to lower values. According to those studies providing soil pH data, pH of representative soil profiles were between 6 and 8 in Hawaii (~20°N) (Dixon et al., 2018), above 5 in the USA (40-45°N) (Ouimet et al., 2015), and between 3 and 5 in the European Alps (45-50°N) (Egli et al., 2010). Hence, some soil-derived ¹⁰Be fluxes in the European Alps might only provide a lower limit. In general, for a given latitudinal band the precipitation-derived fluxes are higher than the soil-derived estimates.

Based on Eq. (3), ¹⁰Be fluxes over a larger spatial scale, that is the river basin-scale, are evaluated. Riverine ¹⁰Be fluxes derived from ¹⁰Be (meteoric)/⁹Be ratios increase with latitude (1° to 30°) and range from 0.24 to 5.74×10⁶ at/cm²/yr, and become quite scattered at 38-40° (the Potomac Basin) with a mean value of 1.84×10⁶ at/cm²/yr. At a higher latitude of ~50° (Czech Republic), riverine ¹⁰Be fluxes decrease to 0.44-1.18×10⁶ at/cm²/yr. Riverine fluxes from the single isotopic system (meteoric ¹⁰Be) are also provided in Supplementary dataset, but are not discussed below.

4. Discussion

4.1. Precipitation collections: dilution or additive effect?

Based on the published precipitation ¹⁰Be dataset and the framework developed in Section 2.5, we can investigate the delivery effect on ¹⁰Be depositional flux over monthly and annual scales. For New Zealand (2 years of observation) and Switzerland (7 years of observation), monthly-measured ¹⁰Be data

are grouped based on sampling seasons and fitted separately (Fig. 5). The slopes are distinct between seasons. The higher slopes in spring and summer are consistent with the seasonal cycles of stratosphere-troposphere exchange, because more 10 Be-enriched stratospheric air is injected into the lower atmosphere in spring and summer (Graham et al., 2003; Heikkilä et al., 2008a). Both in New Zealand and in Switzerland the intercept and the slope show non-zero values, indicating a mixture of additive and dilution components. To derive a representative f_{add} over a certain timescale (seasonal or annual scale), we used the median precipitation rate of all measurements over this period for P in Eq. (4). For New Zealand the resulting f_{add} shows a clear seasonal trend, with lower f_{add} in spring and summer (49-61%) than in autumn and winter (74-87%). For Switzerland, the resulting f_{add} is also lower in spring and summer (59-64%) than in autumn and winter (73-79%). The weaker additive effect in spring and summer can be explained by more frequent occurrence of convective precipitation (dominated by the dilution effect) during this period.

The dataset where ¹⁰Be fluxes integrated from annual records are available can be used to detect the delivery effect over a larger spatial range (Fig. 6). In mid-latitudes (30-50°) a mixture of additive and dilution components exists in islandic (New Zealand) and continental (USA and Switzerland) settings, and the f_{add} are 59% and 62%, respectively. Note that the dilution effect (the slope of the fit line in Fig. 6) cannot be caused solely by dry and recycled ¹⁰Be components. The range of published recycled ¹⁰Be fluxes estimated in the two settings is 0.11-0.33×10⁶ at/cm²/yr (Table S1 in Supplementary materials), and the maximum ¹⁰Be dry depositional flux derived from the preindustrial GCM model run is ~0.19×10⁶ at/cm²/yr (Heikkilä et al., 2013b). As such, the contribution of dry and recycled fluxes (0.30-0.52×10⁶ at/cm²/yr) is much lower than the slope of 0.94-1.00×10⁶ at/cm²/yr in the two settings (Fig. 6). In Kikai Island at ~28° the relatively low slope and comparatively high intercept indicate the dominance of an additive effect, perhaps caused by the high precipitation rate within the observation period. For the dataset at 10-30° (mainly from India), the negative intercept can be explained with F_{prod} (Eq. (A3)) differing

between sampling stations, as this dataset shows a wide range in latitude and a positive correlation $(R^2=0.60)$ between latitude and 10 Be depositional flux.

Overall, our re-analysis of precipitation ¹⁰Be data shows that a mixture of additive and dilution components could be present in both continental and islandic settings. Additionally, the proportion of the additive component seems to decrease with a longer integration timescale. Based on the ECHAM5-HAM output, the ¹⁰Be deposition variability is dominated by precipitation rate over the seasonal scale, but when averaging out the seasonal cycle on multiannual scale, the ¹⁰Be deposition mainly reflects ¹⁰Be production variability rather than precipitation rate (Heikkilä and Smith, 2013), suggesting a larger dilution component and lower fadd. Our analysis is broadly consistent with these modeling results, showing that annual-scale f_{add} (59% and 62%) in both New Zealand and Switzerland is at the lower limit of the seasonal f_{add} range (49-87% and 59-79%, respectively). The potential reason is that the A_c/A_p ratio over multiannual scale may be more stable due to averaging of seasonal variability in vapor sources. Nevertheless, longer time-series precipitation ¹⁰Be records (e.g. decadal-scale) are needed to confirm this temporal variation in f_{add}. In any case, this analysis highlights that the dilution effect does not necessarily dominate global ¹⁰Be precipitation records (Willenbring and von Blanckenburg, 2010b). Also, the empirical equation derived from data fitting between F_{met} and P (Graly et al., 2011) may not result in representative fluxes, because such treatment implicitly assumes a sole additive effect which is mostly not the case.

4.2. Comparing precipitation- with GCM- derived ¹⁰Be fluxes

We next compare ¹⁰Be fluxes determined from precipitation, soil, and river sediment with those derived from GCM modeling (Fig. 7). We do so not because we believe the GCM-derived dataset serves as a benchmark that all other estimates should match, but rather, because of its global spatial coverage making it the only dataset to allow comparison with all other datasets.

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The precipitation-derived fluxes are plotted against GCM-derived fluxes as shown in Fig. 7a. In general, no clear correlation between precipitation- and GCM- derived fluxes is apparent, and the range in precipitation records (0.42-5.30×10⁶ at/cm²/yr) is much larger than that in GCM-derived fluxes (0.66-2.20×10⁶ at/cm²/yr). Almost half of precipitation records are >2 times greater than the corresponding GCM-derived fluxes; 43% of precipitation-derived data are 1-2 times of GCM-derived fluxes, and only 8% of precipitation-derived data is lower than the GCM-derived fluxes (Fig. 7a). This general pattern of significantly higher precipitation-derived fluxes (p < 0.001 from a two-sided Wilcoxon signed-rank test) is independent of location (Fig. 7a). The large variability of precipitation-derived fluxes is consistent with their sensitivity to short-term (10⁰-10¹ yr) local variations in atmospheric dynamics, such as vapor sources and stratosphere-troposphere exchange. Hence, observations of only 1-2 years may not capture the millennial-scale ¹⁰Be depositional signal required for geomorphic applications.

4.3. Comparing soil- with GCM- derived ¹⁰Be fluxes

Over millennial timescale, the relation between soil-derived and GCM-derived fluxes for each latitudinal zone is shown in Fig. 7b. Most of flux data in the "excluded" series yield lower values than other soil data or corresponding GCM-derived fluxes. The potential deficits in ¹⁰Be fluxes can be explained by incomplete sampling of the depth profile or ¹⁰Be loss through surface erosion. For each reliable data series, no correlation between fluxes derived from both methods can be observed, and the range of soil-derived fluxes (0.11-4.07×10⁶ at/cm²/yr) is larger than that of GCM-derived fluxes (1.08-2.55×10⁶ at/cm²/yr). Specifically, GCM-derived data averages over a large grid size (~10² km) and may not fully capture the distinct local variability of ¹⁰Be delivery in Hawaii where precipitation rates increase by one order of magnitude within a short distance (~20 km) (Dixon et al., 2018). The ratio of soil-derived flux to GCM-derived flux is generally lower than 1 (p < 0.05) and varies within a narrow range between 0.48 and 0.90 for average value of each data series. Also, it is worth noting that more than half (57%) of the soil-derived fluxes agree with GCM-derived fluxes within a factor of 2.

4.4. Comparing riverine with GCM-derived ¹⁰Be fluxes

Over the river basin scale, riverine export fluxes derived from ¹⁰Be (meteoric)/⁹Be ratios are compared with GCM-derived meteoric ¹⁰Be fluxes (Fig. 7c). The riverine output of meteoric ¹⁰Be agrees with atmospheric input within a factor of ~2 for each data series, ranging from 0.52 (on average) in the Amazon basin to 1.78 (on average) in the Ganga basin (Fig. 7c). Reasons for a weaker agreement for some samples comprise, for example: 1) coarse-grained quartz used for *in situ* ¹⁰Be measurement (to determine D in Eq. (3)) may be derived from a different sediment source compared to fine-grained sediment used for ¹⁰Be (meteoric)/⁹Be measurement, or multiple grain size fractions analyzed for ¹⁰Be (meteoric)/⁹Be differ in source even at the same location (Wittmann et al., 2015); 2) In basins where no [⁹Be]_{parent} was measured directly and the drainage area is not large enough to average out lithological heterogeneity (e.g. small sub-basins of <100 km² in the Potomac River, USA), the resulting riverine fluxes could be biased by variations in [⁹Be]_{parent}. Nevertheless, although the riverine ¹⁰Be fluxes are generally lower than GCM-derived values (p < 0.001), the majority (71%) of the two estimates indeed fall within a close range (between 2:1 and 1:2 lines) (Fig. 7c).

4.5. Evaluation of ¹⁰Be depositional fluxes from multiple approaches

To provide a more in-depth comparison between different approaches, we selected three regions where both precipitation- and soil- derived ¹⁰Be depositional fluxes exist within a narrow latitudinal band such that both can be compared between each other and with GCM-derived fluxes (Fig. 8). The riverine data is not included as its spatial scale by far exceeds the more local scale over which precipitation or soil ¹⁰Be data integrate. The selected regions include New Zealand (36-46°S), the interior USA (40-45°N) and the European Alps (46-47°N), covering continental and islandic settings. All flux data were normalized to an average Holocene ¹⁰Be production rate.

Over the annual scale, precipitation-derived ¹⁰Be fluxes from the three regions are in close agreement and provide a mean flux of 3.01×10^6 at/cm²/yr. The relative standard deviation of ¹⁰Be fluxes varies from 14-17% (New Zealand and European Alps) to 31% (interior USA). Over the millennial scale, the soil-derived ¹⁰Be fluxes are generally lower than the precipitation-derived ¹⁰Be fluxes (Fig. 8), and amount to

 1.28×10^6 at/cm²/yr on average. On a similar (Holocene) timescale, GCM-derived ¹⁰Be fluxes, with a mean value of 1.76×10^6 at/cm²/yr in the three regions, fall between precipitation- and soil- derived fluxes. Overall, the ratio of average precipitation-derived flux to average GCM-derived flux varies from 1.4 to 1.9. The ratio of average soil-derived flux to average GCM-derived flux falls within a range of 0.5-1.1.

Bearing in mind that all data were normalized to ¹⁰Be production rates with identical solar modulation and magnetic field strength, the fact that ¹⁰Be depositional fluxes from precipitation collections are highest might be caused by a short-term, transient enhancement of ¹⁰Be delivery that is averaged-out over longer timescales (e.g. millennia). Such enhancement may potentially be caused by a transient increase in atmospheric aerosol loading (e.g. from Be scavenging) today due to anthropogenic emission (Dentener et al., 2006). However, this scenario contradicts the findings from GCM modeling, where the difference in GCM-derived fluxes between industrial and early Holocene runs at each site are relatively small (e.g. <~40%), despite of much higher (~5 fold) aerosol loading used in the industrial run (Heikkilä and Smith, 2013). The reason for the low forcing of aerosol loading on ¹⁰Be delivery is that the atmospheric sulfate burden exceeds that of ¹⁰Be by several orders of magnitude under both industrial and pre-industrial conditions (Field et al., 2006; Heikkilä et al., 2013a), and hence ¹⁰Be delivery is production-rather than sulfate-scavenging limited. We can therefore discount this explanation.

Another possibility is that the modern enhancement of ¹⁰Be delivery is a bias introduced through a dependence of precipitation on observational interval. In fact, it was suggested that the precipitation rate depends on measurement interval due to hiatuses (i.e. periods without precipitation) following a power law distribution (Wilkinson, 2015). Over a measurement interval of several years or less, precipitation rate and measurement interval scale by a negative power law, that is, higher precipitation rates are apparent over shorter measurement intervals. This power law behavior emerges from the stochastic nature of synoptic weather systems (Wilkinson, 2015). In contrast, over longer timescales (e.g. decades or longer), the precipitation rate gradually settles to a constant value that reflects the long-term regional climate, and is independent of measurement interval (Wilkinson, 2015). In other words, this measurement

interval bias is not present over a larger spatial or longer temporal span (Sadler and Jerolmack, 2015), such as in GCM and soil profiles, where precipitation rates are deterministic rather than stochastic.

We explore how such measurement interval bias on precipitation rates affects ¹⁰Be delivery. This bias will only affect the additive component of the ¹⁰Be flux because the dilution component does not depend on precipitation rate. Because of the higher precipitation rate over shorter intervals (10⁰-10¹ yr) (Wilkinson, 2015), and if this results from long-distance transport of vapor, the additive ¹⁰Be component f_{add} (Eq. (4)) will increase accordingly and a higher delivery flux of ¹⁰Be will result. Conversely, the measurement interval bias also offers a hypothesis for the lower soil-derived ¹⁰Be fluxes compared to precipitation-derived fluxes: the additive component is suggested to decrease with increasing integration time-interval (Section 4.1) and thus should account for a lower proportion in soil-derived ¹⁰Be fluxes integrating over 10⁴-10⁵ yrs. When corrected for temporal changes of ¹⁰Be production, the dilution component of the ¹⁰Be flux is expected to be time-invariant, and will correspond to the dilution flux contained in precipitation collections. In practice, we can use this assumption to estimate fadd over the timescale integrated by soil profiles. The additive component of the ¹⁰Be depositional flux determined on soils is calculated as the difference between total soil-derived flux and the dilution component of precipitation-derived flux (i.e. slope values in Fig. 6). The resulting proportions of the additive component (f_{add}) for soil profiles in New Zealand (42%) and USA (31%) are indeed much lower than those derived from precipitation records in Fig. 6 (59-62%). In the European Alps the total soil-derived flux (0.76×10^6) at/cm²/yr) is even lower than the precipitation-derived dilution component (1.00×10⁶ at/cm²/yr, Fig. 6). This deficit might hint at a very small additive effect combined with potential loss of soil ¹⁰Be inventory because of the low soil pH (3-5).

The conclusion of these considerations is that, as most geomorphologic processes integrate over millennial scale, ¹⁰Be fluxes from soil profiles rather than precipitation collections provide the most relevant estimate, given their potential to record a representative additive effect over millennia.

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Finally, we explore why GCM-derived fluxes commonly exceed soil-derived fluxes (Fig. 8). One explanation is that soil-derived ¹⁰Be depositional flux provides an underestimate, because the assumption of full ¹⁰Be retention may be invalid (Graly et al., 2011; Maher and von Blanckenburg, 2016). Another explanation is that GCMs provide overestimated fluxes because the production function (Masarik and Beer, 2009) adopted by the GCM (Heikkilä and Smith, 2013) may overestimate (e.g. ~15% higher under a φ of 550 MeV) the global production rate of ¹⁴C (Kovaltsov et al., 2012), and presumably also ¹⁰Be. Since the design of these models, production rates of cosmogenic nuclides by heavier nuclei (atom number ≥ 2) have been revised downwards (Kovaltsov et al., 2012). To conclude, we suggest that the "true" ¹⁰Be depositional flux representative for the millennial scale lies between soil-derived and GCM-derived fluxes reported here.

5. Conclusions

Our re-analysis of ¹⁰Be flux estimates from four independent methods provides important conceptual insights for the use of meteoric ¹⁰Be in Earth surface studies. These are:

- 1) Contemporary ¹⁰Be flux records (precipitation collections) are subject to both a dilution effect and an additive effect. The weight of either effect depends on the area ratio of vapor condensation to precipitation and the prevailing condensation rate. Based on existing precipitation records integrating over the seasonal scale, we estimate the proportion of the additive component to be higher in autumn and winter (up to 87%); over the annual scale, an additive component of ~60% is found in islandic and continental settings. Because only the additive effect depends on precipitation rate, the empirical fitting between ¹⁰Be fluxes and precipitation rates assuming a sole additive effect likely overestimates the control of precipitation on ¹⁰Be delivery.
- 2) ¹⁰Be flux estimates that average over 10³ to 10⁵ years, like those from soil profiles and river fluxes, agree with GCM-derived fluxes within a factor of 2, but can be less than half of the modern ¹⁰Be fluxes from precipitation collections. A possible negative power-law relationship between precipitation rates and

measurement interval over short timescales (10⁰-10¹ yr), combined with a ¹⁰Be additive effect, can explain higher ¹⁰Be fluxes in short-term precipitation collections. Over a longer timescale (10⁴-10⁵ yrs) the additive component can decrease to 30-40%. Because most geomorphologic processes integrate over millennial timescales, ¹⁰Be fluxes from soil profiles, in contrast to precipitation collections, likely provide the most relevant estimate, given their representative estimation of the additive effect over millennia.

3) GCM-derived ¹⁰Be fluxes commonly exceed soil-derived fluxes. The soil-derived fluxes might represent an underestimate due to potential partial retention of ¹⁰Be. GCM on the other hand might overestimate the flux by using outdated spectra of the heavier nuclei component of cosmic rays. We conclude that the best estimate for the millennial-scale ¹⁰Be depositional flux lies between soil- and GCM- derived fluxes.

For producing new ¹⁰Be flux data, we recommend that more efforts should be spent on soil profiles as published data are still rare, and many of these do not meet the stringent requirements for flux calculation. The chosen soil profiles should meet the following criteria: minimum surface erosion, high soil pH, low ¹⁰Be inheritance, and sufficient sampling depth for quantification of the inheritance. If these conditions can be met, ¹⁰Be flux estimates from soil profiles are the method of choice.

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Appendix 1 A mass balance framework for ¹⁰Be delivery effects

The total ¹⁰Be deposition is comprised of wet deposition of ¹⁰Be(OH)₂ scavenged by precipitation, dry deposition of ¹⁰Be(OH)₂ by friction-based aerosol deposition, and recycled ¹⁰Be attached to dust. When neglecting dry and recycled components, we can balance water vapor and ¹⁰Be between vapor

"source" (condensation) and "sink" (precipitation). We conceptually describe the vapor accumulation to be the product of the water condensation rate C ($m^3/m^2/yr$) and the surface area of condensation A_c (m^2), which is balanced by precipitation as calculated by the product of the precipitation rate P ($m^3/m^2/yr$) and the area exposed to precipitation A_p (m^2):

$$C \times A_c = P \times A_p \tag{A1}$$

and thus the precipitation rate P is

$$P = C \times (A_c/A_p) \tag{A2}$$

The ¹⁰Be production flux from the upper atmosphere is F_{prod} (at/m²/yr), and the total flux of ¹⁰Be accumulating in a given moist air mass is proportional to the area A_c where vapor also condenses. The ¹⁰Be concentration of wet deposition ([¹⁰Be]_{wet}, in at/m³) in the condensation area is

$$[^{10}\text{Be}]_{\text{wet}} = (F_{\text{prod}} \times A_c)/(C \times A_c) = F_{\text{prod}}/C$$
(A3)

In the dilution effect, both vapor condensation and 10 Be accumulation take place over the same area as precipitation due to rapid water turnover (e.g. convective precipitation), and thus $A_c = A_p$. Hence, the meteoric delivery flux is:

$$F_{\text{met}} = [^{10}\text{Be}]_{\text{wet}} \times P = F_{\text{prod}}$$
(A4)

In the additive effect, in contrast, moist air mass accumulating both vapor and 10 Be is advected over extended distances (e.g. cyclonic precipitation), and $A_c > A_p$. The meteoric delivery flux is:

$$F_{\text{met}} = [^{10}\text{Be}]_{\text{wet}} \times P = F_{\text{prod}} \times (A_c/A_p)$$
(A5)

Thus, the difference in F_{met} between additive and dilution effects is whether the A_c/A_p ratio (related to the travel distance of moist air mass) controls the precipitation rate (Eq. (A2)) and 10 Be accumulation in moist air mass. Eqs. (A4-A5) form the base for a practical evaluation of the 10 Be delivery effect from combined measurements of 10 Be concentration in precipitation and the precipitation rate.

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656 Figures

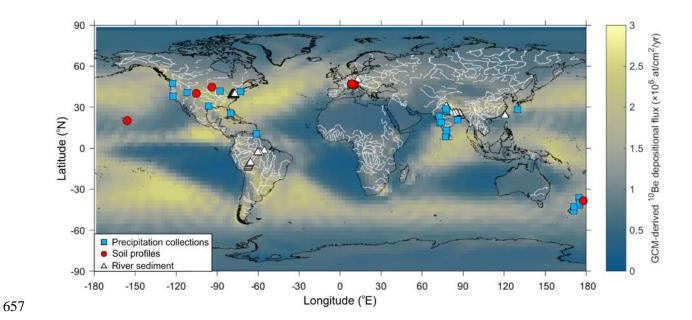


Fig. 1 The spatial distribution of flux data measured in precipitation collections (N=46), soil profiles (N=23) and river sediment (N=90). Data sources are described in detail in Section 2 and the flux data are provided in Supplementary dataset. The distribution map shows the GCM-derived ¹⁰Be depositional flux from the ECHAM5-HAM model with the average ¹⁰Be production and climatic conditions of the Holocene (Heikkilä and von Blanckenburg, 2015).

Normalization of depositional flux

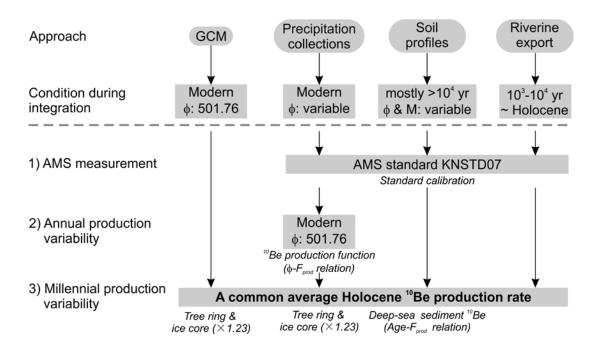


Fig. 2 Procedures for normalization of 10 Be depositional fluxes that account for analytical differences (1) and temporal variations in the flux of galactic cosmic rays (2-3). The aim of these procedures is to normalize all fluxes compiled from four approaches to a common Holocene 10 Be production rate from the upper atmosphere (F_{prod} , in at/ m^2 /yr). ϕ is the solar modulation factor (MeV) and M the geomagnetic field strength. 1) 10 Be concentrations are normalized to a common AMS standard (Nishiizumi et al. (2007). 2) Given that 10 Be is well-mixed in the stratosphere after production (Heikkilä et al., 2013a; Heikkilä et al., 2008b), we perform a global normalization using the modeled relationship between ϕ and global-averaged production rate (Masarik and Beer, 2009). For precipitation collections, each annual depositional flux is normalized using the average ϕ within the observation period to the modern ϕ of 501.76 MeV. 3) Depending on the integration timescale of each approach, fluxes are normalized to an average Holocene production rate using a variety of proxy records of 10 Be production that combine variations of both solar modulation and magnetic field strength (See Fig. S1 in Supplementary materials). The ratio of average Holocene production rate to the modern value at a ϕ of 501.76 MeV (~1.23) is derived from 10 Be production proxy records from tree rings and ice cores (Steinhilber et al., 2012). The global 10 Be

production curve reconstructed from deep-sea sediments over longer term (since 250 ka) is sourced from Christl et al. (2010). The riverine flux estimates were left un-normalized for F_{prod} variations as their integration time scale is unknown and depends on denudation rate and infiltration depth of meteoric 10 Be (Willenbring and von Blanckenburg, 2010b). It is considered to be typically 10^3 - 10^4 yrs, which is within the Holocene reference timescale.

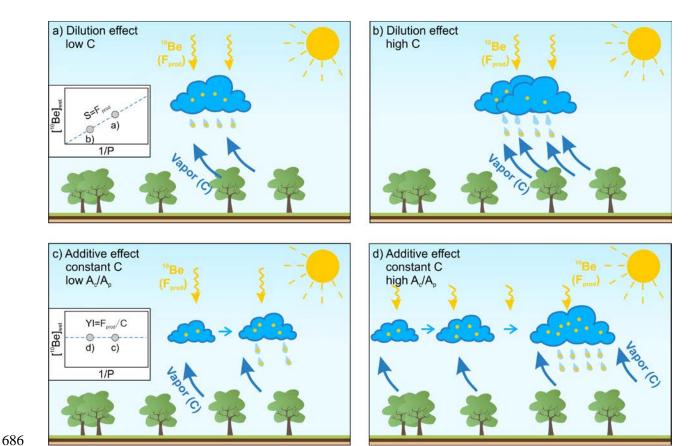


Fig. 3 Conceptual framework of ¹⁰Be delivery. a) and b): the dilution effect is in operation when the ratio of vapor source area (A_c) to area of precipitation event (A_p) is roughly unity, meaning that the vapor is of local origin and also no ¹⁰Be is advected from further distances. In that case the precipitation rate (P, blue droplets) is equal to the condensation rate (C), and the ¹⁰Be flux (yellow dots in droplets) is constant, but the ¹⁰Be concentration depends on the amount of dilution by vapor. The inset in a) shows the relation of [¹⁰Be]_{wet} to 1/P for the dilution effect, and the slope (S, in at/m²/yr) is equal to the atmospheric production rate of ¹⁰Be (F_{prod}). c) and d): the additive effect is in operation when the condensation rate C is constant, but both the precipitation rate P and the amount of ¹⁰Be accumulated scale with A_c/A_p. The inset in c) shows the relation between [¹⁰Be]_{wet} and 1/P for the additive effect (neglecting dry and recycled ¹⁰Be components). Its y-intercept (YI, in at/m³) is set by F_{prod}/C, i.e. the production flux of ¹⁰Be and the water condensation rate. This schematic plot is modified from Willenbring and von Blanckenburg (2010b).

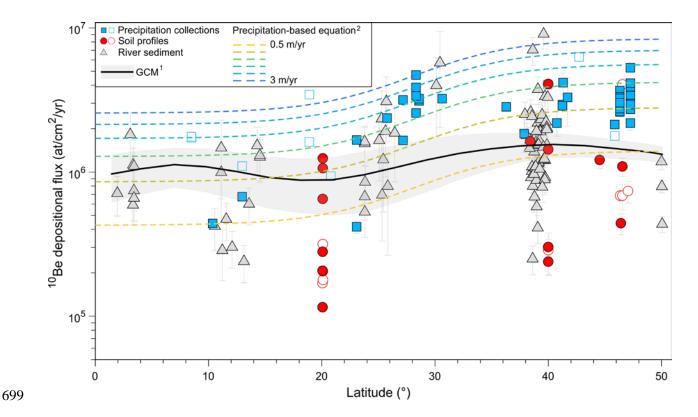


Fig. 4 Latitudinal distribution of measured and modeled ¹⁰Be depositional fluxes derived from the approaches compiled in this study. The latitude shown on the X-axis equally refers to the southern and northern hemispheres, respectively. Note that the latitudes of river sediments are those of sampling locations. 1) GCM results of the average Holocene ¹⁰Be production and climatic conditions are sourced from Heikkilä and von Blanckenburg (2015), and the average flux (black line) and the standard deviation (gray shade) at each latitudinal zone are shown. 2) Results from precipitation-based fitting equation (Eq. (5)) are from Graly et al. (2011). For comparison between different approaches, all flux data were normalized to an average Holocene ¹⁰Be production rate (see Fig. 2). For each dataset, only filled symbols are considered representative and thus used for the discussion of fluxes, whereas flux data shown by open symbols are considered potentially unrepresentative and only shown for completeness.

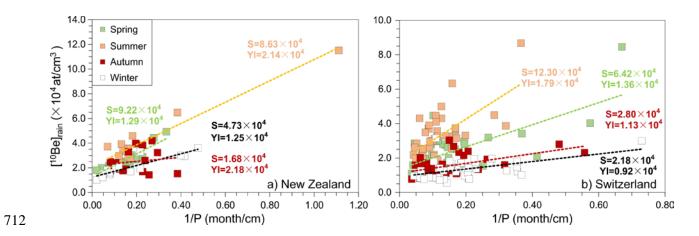


Fig. 5 Plot of the inverse of monthly precipitation rate (1/P) and [10Be]_{rain} in (a) New Zealand and (b)

Switzerland based on monthly records. The slope (S) and y-intercept (YI) are shown for each fit.

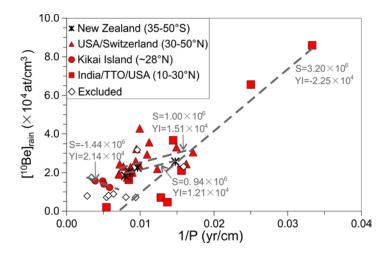


Fig. 6 Plot of inverse of annual precipitation rate (1/P) and mean [10Be]_{rain} based on annual records (data in Supplementary dataset). The slope (S) and y-intercept (YI) are shown for each fit. Series "India/TTO/USA (10-30° N)" includes precipitation data from India (majority), Trinidad (shortened as TTO) and USA at 10-30° N. Note that data from northern India were excluded in a previous data compilation (Graly et al., 2011) as they were assumed to be affected by significant but unquantified dust ¹⁰Be flux. In contrast, we still include such dataset here because recent modeling studies on global dust distribution (Mahowald et al., 2005) show that the dust flux in northern India is not outstandingly high (5 to 20 g/m²/yr). The records with a collection period of less than 9 months are considered unrepresentative and marked as "Excluded" (open symbols). The uncertainties of [10Be]_{rain} are generally lower than the symbol size.

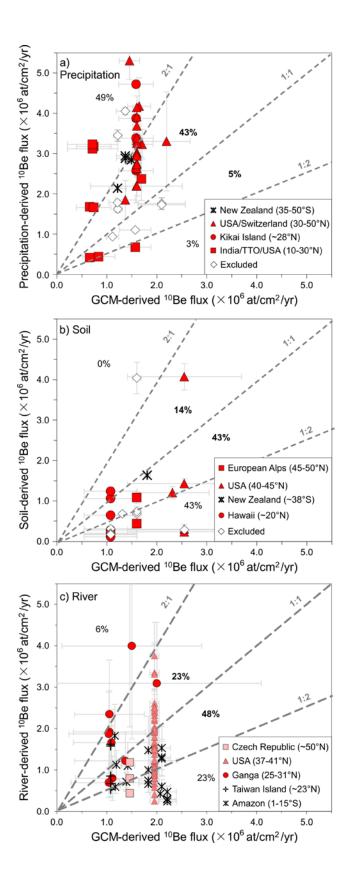


Fig. 7 Comparison between GCM-derived ¹⁰Be depositional fluxes and precipitation- (a) or soil- (b) derived ¹⁰Be depositional fluxes and between basin-averaged ¹⁰Be depositional fluxes from GCM and riverine export ¹⁰Be fluxes (c). Note that error bars of ¹⁰Be fluxes can be smaller than the symbol size for some locations. The GCM-derived flux is based on the average Holocene model of ECHAM5-HAM (Heikkilä and von Blanckenburg, 2015). The corresponding flux uncertainty is the difference between modern and early Holocene model runs, or standard deviation among adjacent grid cell values (applicable for some riverine data, see details in Supplementary dataset). Precipitation- and soil- derived ¹⁰Be flux data are normalized to an average Holocene production rate for comparison. The "Excluded" series in a) is the same as Fig. 6. "Excluded" series in b) includes profiles with significant erosional loss, incomplete sampling or high ¹⁰Be inheritance (see details in Supplementary dataset). Only riverine fluxes derived from ¹⁰Be (meteoric)/⁹Be ratios are shown in (c). Three riverine flux data (range: 5.74-9.09 × 10⁶ at/cm²/yr) are out of the y-axis range and not shown in Fig. 7c, but they are included in the statistics. Each number in the figure represents the percentage of dataset falling in the corresponding domain (divided by the dashed lines).

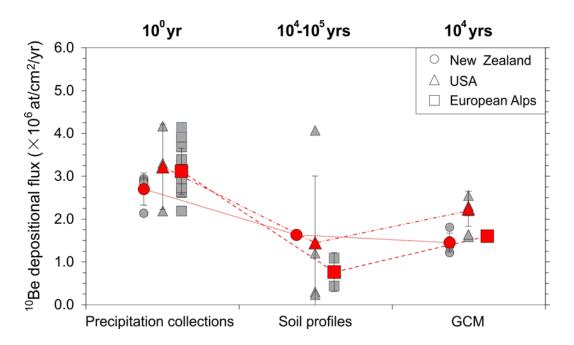


Fig. 8 Comparison of ¹⁰Be depositional fluxes derived from three approaches across timescales. The gray symbols are individual data, and the red symbols are average values derived thereof with an uncertainty of one standard deviation. "Precipitation collections" and "Soil profiles" are measurements over contemporary and millennial timescales, respectively. "GCM" is a modeled dataset derived from ECHAM5-HAM output over average Holocene conditions (Heikkilä and von Blanckenburg, 2015). The integration timescale of each method is provided at the top. All flux data are normalized to the average Holocene ¹⁰Be production rate. Dashed red lines connect mean values of every approach applied in the same region.

1. Reconstruction of paleo-¹⁰Be production rates

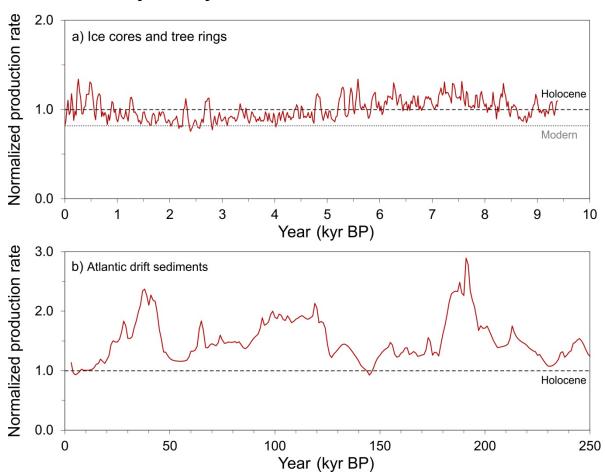


Fig. S1 Temporal variations of the global-averaged ¹⁰Be production rates reconstructed from a) ice cores and tree rings in the Holocene (Steinhilber et al., 2012); b) highly accumulating Atlantic drift sediments since 250 ka (Christl et al., 2010). Each curve is normalized to its average production rate in the Holocene (see the black dashed line). Note that the average ¹⁰Be production rate in the Holocene (a) is ~1.23 times that of the modern value (gray dot line). All ¹⁰Be fluxes in this study are normalized to the average Holocene production rate by dividing by the average normalized production rates during the integration timescale of the archive, upon which the normalization will be done using either curve a) (e.g. for precipitation records) or b) (e.g. for soil profiles).

2. Estimation of recycled ¹⁰Be flux

Table S1 Recycled 10 Be flux estimate in New Zealand, Switzerland and USA. The range is 1.10- 3.28×10^5 at/cm²/yr.

Region	Station	Recycled ¹⁰ Be	Total ¹⁰ Be depositional flux ³	Recycled ¹⁰ Be flux	Data source
		%	at/cm ² /yr	at/cm²/yr	
New Zealand	Gracefield	$9.5\%^{1}$	2.51E+06	2.38E+05	Graham et al. (2003)
New Zealand	Leigh	$9.5\%^{1}$	2.40E+06	2.28E+05	Graham et al. (2003)
New Zealand	Dunedin	16.1% ¹	1.81E+06	2.91E+05	Graham et al. (2003)
Switzerland	Dubendorf	$7.5\%^{1}$	2.31E+06	1.73E+05	Heikkilä et al. (2008)
Switzerland	Jungfraujoch	$6.2\%^{1}$	2.21E+06	1.37E+05	Heikkilä et al. (2008)
USA	Tacoma	$8\%^{2}$	3.58E+06	1.20E+05	Monaghan et al. (1986)
USA	Berkeley	$21\%^{2}$	1.25E+06	1.10E+05	Monaghan et al. (1986)
USA	Salt Lake City	$29\%^{2}$	1.47E+06	1.80E+05	Monaghan et al. (1986)
USA	College Station	$26\%^{2}$	2.17E+06	2.37E+05	Monaghan et al. (1986)
USA	Argonne	$35\%^{2}$	2.23E+06	3.28E+05	Monaghan et al. (1986)
USA	Miami	$26\%^{2}$	1.60E+06	1.75E+05	Monaghan et al. (1986)
USA	New Haven	$20\%^{2}$	2.82E+06	2.37E+05	Monaghan et al. (1986)

Note: 1. The percentage of recycled ¹⁰Be is calculated using ¹⁰Be/¹⁰Be ratios by Graly et al. (2011).

2. The percentage of recycled 10 Be is calculated by multiplying 238 U-derived dust flux by assumed dust 10 Be concentration. However, in the original calculation, Monaghan et al. (1986) assumed an extremely high 10 Be concentration in dust (5×10⁸ at/g) compared to recent observations (2.1×10⁸ at/g on average) (Shen et al., 2010). Hence, we recalculated the recycled 10 Be% (shown here) using a dust 10 Be concentration of 2.1×10^8 at/g.

3. All the ¹⁰Be data are normalized to the AMS standard KNSTD07 (Nishiizumi et al., 2007), but the correction for temporal variability in ¹⁰Be production is not done here. We refer to Table SD1 in Supplementary Dataset for production-normalized fluxes.

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782

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Table SD1 Compiled ¹⁰Be depositional flux data from precipitation collections and the correspon

Station ¹	Country/region	Lat. ²	Lon.	Observation time span			
		°N	°E	Start time	End time		
New Zealand (35-50°	P S)						
Dunedin (1997)	New Zealand	-45,867	170,500	01.10.1997	1998/9/30		
Gracefield (1996)	New Zealand	-41,250	174,917	01.10.1996	30.09.1997		
Gracefield (1997)	New Zealand	-41,250	174,917	01.10.1997	30.09.1998		
Leigh	New Zealand	-36,283	174,800	01.10.1997	30.09.1998		
India, etc (10-30°N)							
Trinidadian	Trinidad	10,361	-61,250	1987	1988		
Bangalore(1980)	India	12,967	77,583	01.07.1980	15.06.1981		
Ahmedabad (1980)	India	23,067	72,633	15.06.1980	04.07.1981		
Ahmedabad (1979)	India	23,067	72,633	22.06.1979	14.06.1980		
Miami	USA	25,757	-80,197	01.08.1980	01.08.1981		
Agra (1980)	India	27,167	78,033	25.06.1980	03.07.1981		
Agra (1979)	India	27,167	78,033	04.07.1979	22.06.1980		
Delhi (1980)	India	28,583	77,200	20.06.1980	10.07.1981		
Delhi (1979)	India	28,583	77,200	14.06.1979	19.06.1980		
Kikai Island (~28°N)							
Site.2 (2000)	Kikai island	28,319	129,985	05.06.2000	24.06.2001		
Site.2 (2001)	Kikai island	28,319	129,985	24.06.2001	29.06.2002		
Site 1 (2000)	Kikai island	28,324	129,928	05.06.2000	24.06.2001		
Site 1 (2001)	Kikai island	28,324	129,928	25.06.2001	29.06.2002		
USA/Switzerland (30	0-50°N)						
College Station	USA	30,628	-96,335	17.10.1980	09.11.1981		
Berkeley	USA	37,871	-122,273	10.12.1980	15.12.1981		
Salt Lake City	USA	40,760	-111,892	04.12.1980	04.12.1981		
New Haven	USA	41,304	-72,929	24.12.1980	07.01.1982		
Argonne	USA	41,706	-87,982	15.09.1980	15.09.1981		
Jungfraujoch (1998)	Switzerland	46,320	7,590	01.01.1998	31.12.1998		
Jungfraujoch (1999)	Switzerland	46,320	7,590	01.01.1999	31.12.1999		
Jungfraujoch (2000)	Switzerland	46,320	7,590	01.01.2000	31.12.2000		
Jungfraujoch (2001)	Switzerland	46,320	7,590	01.01.2001	31.12.2001		
Jungfraujoch (2002)	Switzerland	46,320	7,590	01.01.2002	31.12.2002		
Jungfraujoch (2003)	Switzerland	46,320	7,590	01.01.2003	31.12.2003		
Jungfraujoch (2004)	Switzerland	46,320	7,590	01.01.2004	31.12.2004		
Tacoma	USA	47,248	-122,444	09.08.1980	03.08.1981		
Dubendorf (1998)	Switzerland	47,250	8,270	01.01.1998	31.12.1998		
Dubendorf (1999)	Switzerland	47,250	8,270	01.01.1999	31.12.1999		
Dubendorf (2000)	Switzerland	47,250	8,270	01.01.2000	31.12.2000		
Dubendorf (2001)	Switzerland	47,250	8,270	01.01.2001	31.12.2001		
Dubendorf (2002)	Switzerland	47,250	8,270	01.01.2002	31.12.2002		
Dubendorf (2003)	Switzerland	47,250	8,270	01.01.2003	31.12.2003		
Dubendorf (2004)	Switzerland	47,250	8,270	01.01.2004	31.12.2004		
Below are stations with observation period <9 months							
Dunedin(1997)	New Zealand	-45,867	170,500	01.03.1997	30.09.1997		
Hokitika	New Zealand	-42,717	170,967	01.08.1998	30.11.1998		
Kaitoke	New Zealand	-39,959	175,089	01.01.1997	31.05.1997		
Trivandrium(1980)	India	8,483	76,950	01.04.1980	15.08.1980		

Trivandrium(1979)	India	8,483	76,950	09.07.1979	14.11.1979
Bangalore(1979)	India	12,967	77,583	25.06.1979	19.10.1979
Bombay	India	18,900	72,817	20.06.1979	21.11.1979
Khandala	India	18,900	73,917	23.06.1979	11.08.1979
Cuttack	India	20,800	85,933	21.06.1979	04.11.1979

Note: 1. Stations are sorted and grouped based on latitude. Observations of multiple years at the s

- 2. When coordinates are not provided in the original study (e.g. USA & Trinidad), we estimate La
- 3. Observation period is estimated based on the starting time and end time of sample collection. S
- 4. The ¹⁰Be concentrations used for flux calculation have been normalized to the same AMS standards.
- 5. The average solar modulation factor (φ) during sample collection period is sourced from www.
- 6. All flux data are normalized to a modern ϕ of 501.76 MeV, using the modelled relationship bet
- 7. ϕ -normalized [10 Be]_{rain} is used in Fig. 6.
- 8. The ϕ -normalized flux was further normalized to the average Holocene 10 Be production rate, i.
- 9. The fluxes are calculated based on Eq. (5) in the main text and then normalized to the average
- 10. GCM-derived fluxes for the average Holocene climate conditions are from Heikkilä and von 1

Observation Precipitation		Danasitional		AMS standard-		
	_	Depositional 4	1 sigma	normalized	1 sigma	
period ³	rate	flux ⁴		$[^{10}{ m Be}]_{ m rain}$		
day	cm/yr	at/cm ² /yr	at/cm ² /yr	at/cm ³	at/cm ³	
		J	· · · · · · · · · · · · · · · · · · ·			
365	68	1,81E+06	9,13E+03	2,67E+04	1,35E+02	
365		2,57E+06	9,13E+03	2,20E+04	7,79E+01	
365	104	2,44E+06	9,13E+03	2,35E+04	8,80E+01	
365	126	2,40E+06	1,83E+04	1,91E+04	1,45E+02	
730	184	3,60E+05	0,00E+00	1,95E+03	0,00E+00	
350	78	4,56E+05	3,65E+04	5,85E+03	4,68E+02	
385	73	2,83E+05	2,74E+04	3,87E+03	3,75E+02	
359	64	1,22E+06	6,39E+04	1,91E+04	9,98E+02	
366	118	1,60E+06	3,62E+05	1,35E+04	3,06E+03	
374	64	1,12E+06	2,74E+04	1,75E+04	4,28E+02	
355	30	2,32E+06	9,12E+04	7,72E+04	3,04E+03	
386	69	2,11E+06	6,39E+04	3,05E+04	9,26E+02	
372	40	2,35E+06	7,30E+04	5,88E+04	1,82E+03	
385	253	3,20E+06	6,94E+04	1,26E+04	2,74E+02	
371	202	2,70E+06	5,46E+04	1,34E+04	2,71E+02	
385	195	2,29E+06	9,49E+04	1,17E+04	4,86E+02	
370	170	1,79E+06	3,98E+04	1,06E+04	2,34E+02	
389	115	2,17E+06	2,53E+05	1,89E+04	2,21E+03	
371	61	1,25E+06	3,26E+05	2,03E+04	5,29E+03	
366	58	1,47E+06	2,80E+05	2,53E+04	4,81E+03	
380	105	2,82E+06	4,79E+05	2,68E+04	4,55E+03	
366		2,23E+06	8,32E+05	2,44E+04	9,10E+03	
365	120	2,20E+06	1,73E+05	1,83E+04	1,44E+03	
365	122	2,05E+06	1,73E+05	1,68E+04	1,42E+03	
305		2,49E+06	1,73E+05	1,92E+04	1,33E+03	
335		2,33E+06	1,73E+05	1,65E+04	1,22E+03	
365		2,23E+06	1,73E+05	1,61E+04	1,24E+03	
365		1,87E+06	1,73E+05	1,67E+04	1,54E+03	
365		2,30E+06	1,73E+05	1,89E+04	1,42E+03	
360		3,58E+06	3,16E+05	3,56E+04	3,15E+03	
335		1,84E+06	2,01E+05	2,27E+04	2,49E+03	
305		2,65E+06	2,01E+05	2,49E+04	1,89E+03	
365		2,65E+06	2,01E+05	2,97E+04	2,26E+03	
365		2,90E+06	2,01E+05	2,10E+04	1,46E+03	
365		2,05E+06	2,01E+05	1,82E+04	1,78E+03	
365		1,85E+06	2,01E+05	2,76E+04	3,01E+03	
335	101	2,22E+06	2,01E+05	2,20E+04	1,99E+03	
214		1,57E+06	9,13E+03	2,50E+04	1,45E+02	
122		5,24E+06	2,74E+04	1,78E+04	9,28E+01	
151		3,57E+06	7,30E+04	3,43E+04	7,02E+02	
137	157	1,23E+06	2,74E+04	7,85E+03	1,74E+02	

129	175	1,25E+06	1,00E+05	7,14E+03	5,74E+02
117	124	7,85E+05	3,65E+04	6,33E+03	2,94E+02
155	184	1,16E+06	5,47E+04	6,30E+03	2,98E+02
50	355	2,49E+06	1,00E+05	7,02E+03	2,83E+02
137	106	6,66E+05	2,74E+04	6,26E+03	2,57E+02

same station are separated and the starting time (year) is added behind the station name at. & Lon. from Google Earth.

Samples with observation period< 9 months are not used for statistical analysis in this study. dard KNSTD07.

.faa.gov/data_research/research/med_humanfacs/aeromedical/ radiobiology/heliocentric/ tween ϕ and global-averaged production rate (Masarik and Beer, 2009) as calculated for modern condition

e. multiplied by a factor of ~1.23 according to Steinhilber et al. (2012).

Holocene ¹⁰Be production rates.

Blanckenburg (2015). The dataset is rescaled to an average Holocene production rate.

Average ϕ^5	φ-normalized flux ⁶	1 sigma	φ-normalized [¹⁰ Be] _{rain} ⁷	1 sigma
MeV	at/cm ² /yr	at/cm ² /yr	at/cm ³	at/cm ³
429	1,74E+06	8,77E+03	2,56E+04	1,30E+02
369	2,39E+06	8,46E+03	2,04E+04	7,22E+01
429	2,34E+06	8,77E+03	2,26E+04	8,45E+01
429	2,31E+06	1,75E+04	1,84E+04	1,40E+02
488	3,57E+05	\	1,94E+03	\
907	5,48E+05	4,38E+04	7,02E+03	5,62E+02
901	3,39E+05	3,28E+04	4,64E+03	4,49E+02
719	1,36E+06	7,10E+04	2,12E+04	1,11E+03
911	1,92E+06	4,35E+05	1,63E+04	3,68E+03
907	1,35E+06	3,29E+04	2,11E+04	5,14E+02
719	2,58E+06	1,01E+05	8,59E+04	3,38E+03
907	2,53E+06	7,67E+04	3,67E+04	1,11E+03
721	2,62E+06	8,13E+04	6,55E+04	2,03E+03
904	3,83E+06	8,32E+04	1,51E+04	3,29E+02
830	3,14E+06	6,36E+04	1,56E+04	3,16E+02
904	2,75E+06	1,14E+05	1,41E+04	5,82E+02
830	2,09E+06	4,64E+04	1,23E+04	2,73E+02
924	2,62E+06	3,06E+05	2,28E+04	2,67E+03
913	1,50E+06	3,92E+05	2,44E+04	6,37E+03
921	1,78E+06	3,38E+05	3,05E+04	5,81E+03
909	3,39E+06	5,76E+05	3,22E+04	5,46E+03
909	2,68E+06	1,00E+06	2,93E+04	1,09E+04
446	2,13E+06	1,67E+05	1,78E+04	1,40E+03
578	2,14E+06	1,80E+05	1,75E+04	1,47E+03
909	2,99E+06	2,07E+05	2,31E+04	1,60E+03
819	2,70E+06	2,00E+05	1,91E+04	1,42E+03
868	2,64E+06	2,04E+05	1,90E+04	1,47E+03
903	2,24E+06	2,07E+05	2,00E+04	1,85E+03
637	2,46E+06	1,85E+05	2,02E+04	1,52E+03
911	4,31E+06	3,81E+05	4,29E+04	3,79E+03
446	1,78E+06	1,95E+05	2,20E+04	2,41E+03
578	2,75E+06	2,10E+05	2,59E+04	1,97E+03
909	3,18E+06	2,42E+05	3,57E+04	2,71E+03
819	3,36E+06	2,34E+05	2,43E+04	1,69E+03
868	2,43E+06	2,38E+05	2,15E+04	2,11E+03
903	2,22E+06	2,41E+05	3,31E+04	3,60E+03
642	2,39E+06	2,16E+05	2,36E+04	2,14E+03
360	1,45E+06	8,42E+03	2,30E+04	1,34E+02
459	5,12E+06	2,68E+04	1,74E+04	9,07E+01
360	3,29E+06	6,73E+04	3,16E+04	6,47E+02
774	1,40E+06	3,12E+04	8,94E+03	1,99E+02

781	1,43E+06	1,15E+05	8,16E+03	6,55E+02
781	8,96E+05	4,17E+04	7,23E+03	3,36E+02
768	1,32E+06	6,22E+04	7,15E+03	3,38E+02
742	2,80E+06	1,13E+05	7,88E+03	3,18E+02
781	7,61E+05	3,13E+04	7,15E+03	2,94E+02

ıs (Heikkilä et al., 2013b).

Production-		Precipitation data	GCM-derived	
normalized flux ⁸	1 sigma	fitting-based ¹⁰ Be flux ⁹	flux (average Holocene) ¹⁰	1 sigma
at/cm ² /yr	at/cm ² /yr	at/cm ² /yr	at/cm ² /yr	at/cm ² /yr
	•	·	·	<u>, </u>
2,14E+06	1,08E+04	1,86E+06	1,22E+06	1,70E+05
2,94E+06	1,04E+04	3,11E+06	1,37E+06	1,80E+05
2,88E+06	1,08E+04	2,75E+06	1,37E+06	1,80E+05
2,84E+06	2,16E+04	3,00E+06	1,49E+06	1,80E+05
4,40E+05 \		1,61E+06	8,35E+05	3,24E+05
6,75E+05	5,40E+04		1,57E+06	3,14E+05
4,17E+05	4,04E+04	•	6,55E+05	3,22E+04
1,67E+06	8,75E+04	•	6,55E+05	3,22E+04
2,37E+06	5,36E+05	1,57E+06	1,69E+06	3,14E+03
1,66E+06	4,05E+04		7,36E+05	3,44E+05
3,17E+06	1,25E+05	*	7,36E+05	3,44E+05
3,12E+06	9,44E+04	•	7,14E+05	5,04E+05
3,23E+06	1,00E+05	*	7,14E+05	5,04E+05
4.72E+06	1.020.05	2.095+06	1.500 +06	2.45E+05
4,72E+06	1,02E+05	3,98E+06	1,58E+06	3,45E+05
3,87E+06	7,84E+04	•	1,58E+06	3,45E+05
3,38E+06	1,40E+05	·	1,58E+06	3,45E+05
2,57E+06	5,71E+04	2,67E+06	1,58E+06	3,45E+05
3,23E+06	3,77E+05	2,09E+06	1,71E+06	2,12E+05
1,85E+06	4,82E+05	1,53E+06	1,37E+06	1,99E+05
2,19E+06	4,17E+05	1,53E+06	1,59E+06	9,20E+05
4,17E+06	7,09E+05	2,80E+06	1,65E+06	2,30E+05
3,30E+06	1,23E+06	·	2,20E+06	4,70E+05
2,62E+06	2,06E+05	3,31E+06	1,60E+06	8,00E+04
2,63E+06	2,21E+05	3,36E+06	1,60E+06	8,00E+04
3,68E+06	2,55E+05	3,57E+06	1,60E+06	8,00E+04
3,33E+06	2,47E+05	3,89E+06	1,60E+06	8,00E+04
3,25E+06	2,51E+05		1,60E+06	8,00E+04
2,76E+06	2,55E+05	3,09E+06	1,60E+06	8,00E+04
3,03E+06	2,28E+05	3,36E+06	1,60E+06	8,00E+04
5,30E+06	4,69E+05	2,78E+06	1,45E+06	2,03E+05
2,19E+06	2,40E+05	•	1,60E+06	7,88E+04
3,39E+06	2,58E+05		1,60E+06	7,88E+04
3,91E+06	2,98E+05	2,47E+06	1,60E+06	7,88E+04
4,14E+06	2,88E+05	3,83E+06	1,60E+06	7,88E+04
2,99E+06	2,93E+05	3,13E+06	1,60E+06	7,88E+04
2,73E+06	2,97E+05	1,86E+06	1,60E+06	7,88E+04
2,94E+06	2,66E+05	2,80E+06	1,60E+06	7,88E+04
1,78E+06	1,04E+04	1,73E+06	1,21E+06	1,75E+05
6,30E+06	3,30E+04		1,26E+06	2,25E+05
4,05E+06	8,29E+04	•	1,37E+06	1,81E+05
.,	-,	_, 00	-,	-,-12.30

1,76E+06	1,41E+05	1,52E+06	2,10E+06	4,73E+05
1,10E+06	5,13E+04	1,10E+06	1,57E+06	3,12E+05
1,62E+06	7,66E+04	1,80E+06	1,22E+06	8,40E+04
3,45E+06	1,39E+05	3,47E+06	1,22E+06	8,40E+04
9,37E+05	3,85E+04	1,11E+06	1,11E+06	1,24E+05

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