A global dataset of atmospheric ^{7}Be and ^{210}Pb measurements: annual air concentration and depositional flux

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Abstract. ^{7}Be and ^{210}Pb air concentration and depositional flux data provide key information on the origins and movements of air masses, as well as atmospheric deposition processes and residence time of aerosols. After their deposition onto the Earth' surface, they are utilized for tracing soil redistribution processes on land, particle dynamics in aquatic systems and mixing processes in open ocean. Here we present a global dataset of air concentration and depositional flux measurements of atmospheric ^{7}Be and ^{210}Pb made by a large number of global research community. Data were collected from published papers between 1955 and early 2020. It includes the annual surface air concentration data of ^{7}Be from 367 sites and of ^{210}Pb from 270 sites, the annual depositional flux data of ^{7}Be from 279 sites, and of ^{210}Pb from 602 sites. When available, appropriate metadata have also been summarized, including geographic location, sampling date, methodology, annual precipitation, and references. The dataset is archived at https://doi.org/10.5281/zenodo.4785136 (Zhang et al., 2021) and is freely available for the scientific community. The purpose of this paper is to provide an overview of the scope and nature of this dataset and its potential utility as baseline data for future research.

1 Introduction

Naturally occurring beryllium-7 (^{7}Be, $T_{1/2}$: 53.3 days) and lead-210 (^{210}Pb, $T_{1/2}$: 22.3 y) have been widely utilized as tracers to investigate Earth' surface and atmospheric processes (Huh et al., 2006; Du et al., 2012). ^{7}Be, a cosmogenic radionuclide, is produced by the spallation of oxygen and nitrogen nuclei by cosmic rays in the stratosphere and upper troposphere (Lal et al., 1958). The production rate of ^{7}Be has negligible dependence on longitude or season, but depends on the altitude, latitude and the ~11 years solar cycle (Koch et al., 1996; Liu et al., 2001; Su and Huh, 2003). A major fraction of ^{7}Be (67%) production takes place in the stratosphere, but it does not readily reach the troposphere except during spring when seasonal thinning of the tropopause folds near the jet stream occurs at mid-latitudes (Lal and Peters, 1967; Danielsen, 1968). Thus, ^{7}Be flux to the Earth' surface varies with latitude and season (Lal and Peters, 1967; Koch and Mann, 1996). ^{210}Pb, a progeny of ^{222}Rn in the ^{238}U-series, is derived mostly (>99%) from the radioactive decay of ^{222}Rn. Most of the atmospheric ^{210}Pb is derived from atmospheric radon. The global ^{222}Rn flux from continent ranged from 1300 to 1800 Bq m^{-2} d^{-1}, while 2-21 Bq m^{-2} d^{-1} were reported for the oceanic areas (Wilkening and Clements, 1975; Nazaroff, 1992). Vertical profiles of ^{222}Rn in the atmosphere indicate the highest concentrations occur in the continental boundary layer (CBL, 3-8 Bq m^{-3}) while an order of magnitude lower activity (~40 mBq m^{-3}) occurs near the tropopause, with decreasing activity with increasing altitude from CBL (Moore et al., 1977; Liu et al., 1984; Kritz et al., 1993). Consequently, the atmospheric concentration of ^{210}Pb decreases with increasing altitude and is strongly controlled by the land-sea distribution pattern. After formation, both ^{7}Be and ^{210}Pb and short-lived ^{222}Rn progeny, are rapidly and irreversibly attached to aerosol particles (Winkler et al., 1998; Elsässer et al., 2011). Subsequently, the fate of ^{7}Be and ^{210}Pb is closely linked to that of aerosols. Most of ^{7}Be and ^{210}Pb are in accumulation mode aerosol particles with an aerodynamic diameter of a few hundred nanometers (Ioannidou and Paatero, 2014; Paatero et al., 2017). Therefore, they are deposited onto the Earth' surface primarily by precipitation because accumulation mode aerosol particles are too small for gravitational settling and removal and too large to be deposited by Brownian motion.

Owing to their distinctly different source terms but well-known source distributions, and similar tropospheric physicochemical behavior, ^{7}Be and ^{210}Pb have been widely utilized as powerful atmospheric tracers for studying the origin of air masses (e.g., Graustein and Turekian, 1996; Zheng et al., 2005; Likuku et al., 2006; Dueñas et al., 2011; Lozano et al., 2012), vertical exchange and horizontal transport processes (e.g., Arimoto et al., 1999; Lee et al., 2007; Rastogi and Sarin, 2008; Tositti et al., 2014), deposition velocities and washout ratios of aerosols (e.g., Todd et al., 1989; McNeary and Baskaran, 2003, Dueñas et al., 2005; Lozano et al., 2011; Mohan et al., 2019) and behavior and fate of analog species (e.g., Crecelius, 1981; Mattson, 1988; Prospero et al., 1995; Lamborg et al., 2013). Following their deposition on the Earth' surface, both ^{7}Be and ^{210}Pb are strongly attached to soils, which make them useful for assessing soil erosion rates from episodic to multi-decadal timescales (e.g., Wallbrink and Murry, 1993; Walling and He, 1999; Blake et al., 1999; Walling et al., 1999; Wilson et al., 2003; Mabit et al., 2008, 2014). In aquatic environments, ^{210}Pb is most widely used for dating recent sediments (e.g., Appleby, 2008). Meanwhile, ^{7}Be and ^{210}Pb are also widely used as tracers of sediment source identification and particle dynamics in rivers (e.g., Bonniwell et al., 1999; Matisoff et al., 2005; Jweda et al., 2008; Mudbidre et al., 2014; Baskaran et al., 2020), lakes (e.g., Dominik et al., 1987; Schuler et al., 1991; Vogler et al., 1996), estuaries and coasts (e.g., Baskaran et al., 1997; Huang et al., 2013; Wang et al., 2016). ^{7}Be deposited on open ocean is further used as a tracer for diagnosing ocean ventilation and subduction (Kadko, 2000; Kadko and Olson, 1996), inferring upwelling rates (Kadko and Johns, 2011) and estimating the deposition of trace metals (Kadko et al 2015; Shelley et al., 2017; Buck et al., 2019).

There have been numerous published datasets with concentrations and depositional fluxes of ^{210}Pb and ^{7}Be (directly and indirectly) over the past few decades, particularly under the national or international monitoring programs such as Environmental Measurements Laboratory (EML) Surface Air Sampling Program (Feely et al., 1989), Sea-Air Exchange (SEAREX) program (Uematsu et al., 1994), Finnish Meteorological Institute monitoring program (Paatero et al., 2015), Radioactivity Environmental Monitoring (REM) network (Hernandez-Ceballos et al., 2015), and International Monitoring System (IMS) operated by Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) (Terzi and Kalinowski, 2017). It is valuable to compile all these existing data, including those measured in case studies, along with appropriate metadata, in one place for facilitating further data analysis and geological, geochemical and geophysical applications.

Although several datasets of air concentrations and depositional fluxes of ^{7}Be (Bleichrodt, 1978; Brost et al., 1991) and ^{210}Pb (Rangarajan et al., 1986; Preiss et al., 1996) have been published, unfortunately, many of the published data are not readily available in hard data format (e.g., data table) and often it is challenging and retrieval of data from published figures often is not precise. To date, only one dataset was published that compiled ^{7}Be and ^{210}Pb together (Persson, 2015), but it contained limited data. Therefore, the focus of the present work is to build a new and comprehensive dataset. This dataset is the result of many scientists' efforts in generating the data.

2 Methods

2.1 ^{7}Be and ^{210}Pb air concentrations measurement methodology

Aerosol samples are usually collected by filtering a high volume of air, typically 1.4-1.5 m^3/min, collected typically for a day, through paper filters. Common aerosol collecting equipment includes ASS 500 (CLRP Warsaw, Germany), Anderson PM10 (Anderson Ltd., USA), Snow White (Senya Ltd., Finland), and HV-1000F (Shibata Co. Ltd., Japan). The preferred filter membrane material includes glass fiber, cellulose nitrate or acetate, polypropylene fiber, and quartz fiber. The collection efficiency, the percentage of the particles in the air stream that are collected by the air filter, depends on the aerodynamic diameter of aerosol particles and the filter face velocity (average flow velocity of air into the filter) of the airflow. Although the collection efficiency depends on the distribution of particle sizes, generally the collection efficiency varies between 80% and 100% for different filter materials. This was shown by overlapping two filters in tandem and comparing the concentrations separately in the top and bottom filters. The sampling frequency is usually set from daily to monthly, with a typical collection time of ~ 24 h, corresponding to ~ 2000 m^3 air. The ^{7}Be and ^{210}Pb trapped on filters can both be analyzed simultaneously by gamma spectrometry (e.g., McNeary and Baskaran, 2003; Bourcier et al., 2011; Lozano et al., 2012; Mohan et al., 2018), while ^{210}Pb can also be analyzed by beta counting of its daughter ^{210}Bi (e.g., Joshi et al., 1969; Poet et al., 1972; Daish et al., 2005) or via alpha counting of in-grown ^{210}Po from the decay of ^{210}Pb (e.g., Turekian and Cochran, 1981; Mattsson et al., 1996; Marx et al., 2005).

2.2 ^{7}Be and ^{210}Pb depositional fluxes measurement methodology

The atmospheric depositional fluxes of ^{7}Be and ^{210}Pb are commonly measured directly by using rain collectors such as polyethylene drum/buckets, stainless steel container, and indirectly by natural archives (e.g., soils, lichens, mosses, snow/ice cores, salt marsh sediments). The direct collecting method is the most reliable technique for the measurement of annual ^{7}Be and ^{210}Pb depositional fluxes, and this technique is useful in collecting short-time scale (daily, weekly and monthly) depositional fluxes. In contrast, using natural archives avoids the labor and time-intensive measurement of the ^{7}Be and ^{210}Pb concentration in precipitation and can serve as a complement to fill regional gaps especially in remote areas. However, these archives are susceptible to be affected by natural processes and anthropogenic activities, thus, the sampling location of these archives should be restricted to undisturbed areas.

2.2.1 Direct ^{7}Be and ^{210}Pb flux measurements

Rain collectors are usually placed on the roof of a building so as to prevent contamination from resuspended dust from the ground. Care should be taken to ensure direct overhead atmospheric deposition is collected and there is no shadowing effect from adjacent structure/building or funneling effect in sample collection. Atmospheric aerosols can be removed not only by precipitation-scavenging but also by settling under the influence of gravitational or electrostatic forces. In most cases, the collectors are continuously exposed over a long enough period and the bulk (wet + dry) depositional samples are collected periodically (e.g., Baskaran et al., 1993; Hirose et al., 2004; Baskaran and Swarzenski, 2007; Lozano et al., 2011; Du et al., 2015). Fluxes obtained by this method yield the best estimate of the depositional flux. Sometimes, fallout ^{7}Be and ^{210}Pb samples are collected only during rainfall, and concentration is measured in the individual rainwater sample (e.g., Cho et al., 2011; Chae and Kim, 2019; Du et al., 2020). In this case, only the bulk depositional flux is obtained for the duration of collection. In rare cases, only a mean concentration of ^{210}Pb and/or ^{7}Be in rainwater is available, the wet flux can be estimated by multiplying by the annual precipitation (Peirson et al., 1966).

Most of the time, the volume of rainwater sample is large which cannot be directly counted in a gamma-ray spectrometer for the simultaneous measurements of ^{7}Be and ^{210}Pb, in which case a preconcentration of the sample is required. Since both ^{7}Be and ^{210}Pb have a strong affinity for solid surfaces, it is strongly recommended to add stable Be (commonly 1-5 mg) and stable Pb (typically 5-20 mg) in about 1 L of 1 M HCl to the rain collector prior to deployment. Alternatively, the spikes can also be immediately added after sample collection, followed by rinsing of rain collector with 1 L of 1 M HCl rinsing twice and combining the rinses with the collected rainwater. An earlier critical review of earlier atmospheric depositional flux studies by Lal et al. (1979) showed that loss of ^{7}Be and ^{210}Pb by sorption onto rain collector walls was observed when pre-acidification of the collector was not done resulting in the underestimation of depositional flux. In the case of preconcentration by ferric chloride precipitation method, due to variable scavenging efficiency of ^{7}Be and ^{210}Pb during the preconcentration method, it is required to add stable Pb and Be as yield tracer (e.g., Baskaran et al., 1993). The best chemical procedure to obtain high-quality data is to add acid and stable Be and Pb careers with 1 L of 1 M HCl to the rain collector prior to the start of the sample collection. The final calculation for depositional fluxes would involve chemical yield for ^{7}Be and ^{210}Pb, and appropriate decay corrections, as outlined in Baskaran et al. (1993) and Du et al. (2015).

2.2.2 Indirect ^{7}Be flux measurements

Measurements of ^{7}Be inventory in the upper oceanic water column, from air-sea interface until the layer where ^{7}Be activity is below the detection limit (in >400 L water sample), can indirectly yield the bulk depositional flux of ^{7}Be (Brost et al., 1991). This requires precise determination of the penetration depth of ^{7}Be in the water column. The only uncertainty is the loss of ^{7}Be-laden sinking particles from the upper water column where ^{7}Be is present. After being deposited on the ocean surface, ^{7}Be is generally mixed uniformly within the surface mixed layer (Young and Silker, 1980; Kadko and Olson, 1996). In open ocean, the particle concentration is generally low, and a major fraction of ^{7}Be is expected to be in the dissolved phase, thus allowing particle scavenging losses to be ignored (Silker, 1972; Andrews et al., 2008). Therefore, in the absence of physical removal processes other than radioactive decay, the input flux of ^{7}Be should be balanced by the ^{7}Be inventory integrated over the water column. In other words, ^{7}Be flux of atmospheric fallout (Bq m^{-2} d^{-1}) can be obtained from the ^{7}Be water column inventory (Bq m^{-2}) multiplied by the decay constant (0.013 d^{-1}) of ^{7}Be. This method has been proven to be reliable in open ocean due to the relatively short half-life of ^{7}Be and the constancy of ^{7}Be deposition over broad latitudinal bands (Young and Silker, 1980; Aaboe et al., 1981). It is expected that the ^{7}Be inventory is season-dependent in areas with large seasonal variations in precipitation (e.g., monsoon-dominated continental and oceanic areas). Time-series study in Bermuda has shown that the inventory of ^{7}Be was relatively constant throughout the year, such that ^{7}Be inventory measured at any one time is likely representative (to within 20%) of the instantaneous ^{7}Be flux (Kadko and Prospeo, 2011; Kodko et al., 2015). This method is not suitable for coastal and estuarine areas where ^{7}Be is scavenged substantially by particulate matters (Olsen et al., 1986; Baskaran and Santschi, 1993), and upwelling-dominated areas where ^{7}Be inventory is diluted by ^{7}Be "dead" water (Kadko and Johns, 2011; Haskell et al., 2015).

Another potential candidate is undisturbed soil profiles. However, ^{7}Be inventories in undisturbed soils were reported to vary by more than an order of magnitude within one year (Walling et al., 2009; Kaste et al., 2011; Zhang et al., 2013). Such large variations in depositional fluxes of ^{7}Be are attributed to the seasonal fluxes of ^{7}Be. Earlier studies have shown that the atmospheric fluxes are highly dependent upon the amount of precipitation (Baskaran, 1995; Du et al., 2015). Since seasonal variations will significantly affect the ^{7}Be inventory in soils, the data of ^{7}Be soil inventory are not included in our dataset.

2.2.3 Indirect ^{210}Pb flux measurements

Several archives (soils, snow/ice cores, sediment cores, etc) have been used to assay the ^{210}Pb fluxes. Here we only present ^{210}Pb fluxes estimated from soil profiles and snow/ice cores. The former is the most frequently used, and the latter perfectly fills the regional gap in polar regions and montane permanent snowfields. There are many ^{210}Pb measurements in sediment cores, however, due to the sediment focusing and erosion, most sediment cores do not provide a reliable estimate of the atmospheric ^{210}Pb flux (Turekian et al., 1977; Preiss et al., 1996), thus this type of ^{210}Pb depositional flux data are not included in our dataset.

^{210}Pb in surface (upper ~30 cm) soil has two sources: one is generated from the decay of ^{222}Rn in the soil minerals, known as supported ^{210}Pb which is produced from the decay of ^{238}U and the other comes from atmospheric deposition as unsupported ^{210}Pb. The fallout of ^{210}Pb is retained generally in the organic-rich surface soils presumably because of the sequestering properties of the organo-mineral complexes (Covelo et al., 2008). When soil CO_2 combines with percolating water, carbonic acid is produced which can leach some of the sorbed ^{210}Pb ultimately resulting in slow migration down to a depth of up to 20-30 cm (Matisoff and Whiting, 2012). As a result, the surface soil layer contains an excess ^{210}Pb compared to that from its equilibrium with ^{226}Ra (Mabit et al., 2014). The part of the ^{210}Pb excessing is termed "unsupported" or "excess" ^{210}Pb ($^{210}Pb_{ex}$). $^{210}Pb_{ex}$ is the difference between total (measured) ^{210}Pb and the supported ^{210}Pb in the soils. Supported ^{210}Pb is assumed to be the same as ^{226}Ra activity, under the assumption of secular equilibrium between ^{226}Ra and supported ^{210}Pb. It can also be obtained by assuming that the supported ^{210}Pb activity is equal to the total ^{210}Pb at depth greater than 30 cm in the soil profile where atmospherically-delivered ^{210}Pb has not reached (Matisoff et al., 2014). The mean residence time of ^{210}Pb over a large drainage basin is on the order of 2000-3000 years in surface soils (Benninger et al., 1975; Dominik et al., 1987), so the inventory of $^{210}Pb_{ex}$ in a soil profile that has not been disturbed by erosion, accumulation or human activities for about a century can be used to calculate the depositional flux (Graustein and Turekian, 1986). At steady state, the ^{210}Pb depositional flux can be deduced using the $^{210}Pb_{ex}$ inventory multiplied by the decay constant (0.0311 y^{-1}) of ^{210}Pb. This method has been widely used worldwide (e.g., Nozaki et al., 1978; Graustein and Turekian, 1986, 1989; Dörr and Munich, 1991; García-Orellana et al., 2006). At undisturbed soil sites, flux values derived from soil profile measurements were consistent with direct atmospheric flux observations (Olsen et al., 1985; Appleby et al., 2002, 2003), and $^{210}Pb_{ex}$ soil inventory showed little discrepancy at different sampling times (Porto et al., 2006, 2016).

Goldberg (1963) was the first to show that the total ^{210}Pb activity in a glacier from Greenland decreased with depth, with a possibility of dating ice cores. Subsequently, snow chronology in the Antarctic was determined (Crozaz et al. 1964; Picciotto et al., 1964). Since then, this technique has been used in both the large ice caps of Antarctica (e.g., Picciotto et al., 1968; Koide et al., 1979; Nijampurkar et al., 2002) and the Arctic (e.g., Crozaz and Langway, 1966; Koide et al., 1977; Dibb et al., 1990a, 1992; Peters et al., 1997) and the small montane permanent snowfield (e.g., Windom, 1969; Gäggeler et al., 1983; Monaghan and Holdsworth, 1990). The ^{210}Pb flux in snow/ice core is calculated in the same way as for the soil, except that supported ^{210}Pb in snow/ice core is very low due to low concentration of ^{226}Ra in snow/ice core and may be negligible, as the lithogenic dust is the primary source of ^{226}Ra and its concentration in polar regions is very low (Preiss et al., 1996). When the snow accumulation rate is known, the depositional flux can also be obtained by using ^{210}Pb concentration in surface snow multiplied by the accumulation rate (Pourchet et al., 1997; Suzuki et al., 2004). The uncertainty in the depositional flux of ^{210}Pb from snow/ice core record is the potential post-depositional movement of the snow/ice due to heavy wind and the possibility of snow melting and percolation.

2.3 Data collection

In order to compile the global dataset for annual ^{7}Be and ^{210}Pb air concentrations and depositional fluxes comprehensively, we attempted to collect published papers between 1955 and early 2020 in which hard data for their concentrations and depositional fluxes are available or their calculated values reported. Using a series of keywords or with a combination of words search (e.g., ^{7}Be, ^{210}Pb, air concentration, depositional flux, fallout radionuclide, atmospheric tracer, or soil erosion), data were retrieved from online literature databases (Web of Science, Science Direct, and China Knowledge Resource Integrated Database). During the literature survey, no a priori criteria (e.g., study area, sampling period, and measurement method) were applied. However, a critical review of the collected literature was conducted to obtain long-term data, using the following criteria. For concentrations in air and directly measured fluxes of ^{7}Be and ^{210}Pb, only those sites with more than one year of data were included. When averaged over a longer period, data are more representative because of the inherent seasonal variations (at least one full year data) and inter-annual fluctuations (multi-year data). For indirectly measured fluxes of ^{7}Be

and ^{210}Pb, only those undisturbed sites clearly stated in the original literature were included.

Here we did not include unpublished data, as data quality control could be a potential issue. In the peer-reviewed published data, it is assumed that the authors, reviewers and/or editors of the original articles have undertaken the necessary steps to verify data quality. All concentrations in air were converted into mBq m^{-3} and all depositional fluxes were converted into Bq m^{-2} y^{-1}, if not already reported in these units. When available, the metadata of latitude, longitude, altitude, sampling date, annual precipitation, methodology, and references are also given. A brief description of different variables that could affect the data is summarized in Table 1. In cases where the air concentration and depositional flux data were only available graphically, a computer program (GetData Graph Digitizer) was used to digitize the data from graphics, the same was done for geographical location and annual precipitation. In rare cases, only the locality name of the study site was available, the geographical coordinates were extracted from Google Earth.

Table 1. Fields in the main data table.

Field name	Field description	Field type	Unit
Site	Locality name (city, country) or station name of study site	Short text	Unitless
Sampling time	Monitoring period or sampling date in month/year format	Short text	Unitless
Latitude	North latitude in decimal degrees (from -90 to +90) either directly from original studies or extracted from Google Earth	Number	°
Longitude	East longitude in decimal degrees (from -180 to +180) either directly from original studies or extracted from Google Earth	Number	°
Altitude	Altitudes of study sites either directly from original studies	Number	m
Annual precipitation	Mean annual precipitation of study sites directly from original studies	Number	mm
Sampling device[a]	Model of aerosol sampling device and air flow rate during sampling	Short text	Unitless
Filter[a]	Model, material and dimension of filter membranes	Short text	Unitless
Frequency[a]	Sampling frequency set during observation	Short text	Unitless
Data number[a]	Total number of measurements performed	Integer	Unitless
Concentration[a]	^{7}Be or ^{210}Pb annual concentration in surface air	Number	mBq m^{-3}
Con-error[a]	Uncertainty in ^{7}Be or ^{210}Pb annual concentration in surface air	Number	mBq m^{-3}
Con-range[a]	Minimum to maximum values of ^{7}Be or ^{210}Pb concentration	Number	mBq m^{-3}
Method[b]	Short description of ^{7}Be or ^{210}Pb depositional flux obtained from different methods	Short text	Unitless
Rain collector[b]	Material, shape and collection area of rain collectors	Short text	Unitless
Clean procedure[b]	Rainwater collector cleaning process and reagents used	Short text	Unitless
Preconcentration[b]	Preconcentration method for rainwater samples	Short text	Unitless

Recovery[b]	Recovery of ^{7}Be or ^{210}Pb and the determination method	Short text	Unitless
Decay-correction[b]	Calculation of decay-correction for ^{7}Be or ^{210}Pb	Short text	Unitless
Flux[b]	^{7}Be or ^{210}Pb annual depositional flux derived from different methods	Number	Bq m^{-2} y^{-1}
Flux-error[b]	Uncertainty in ^{7}Be or ^{210}Pb annual depositional flux	Number	Bq m^{-2} y^{-1}
Contribution of dry deposition[b]	Fraction of dry depositional flux to total depositional flux of ^{7}Be or ^{210}Pb	Number	Unitless
Reference	Investigator and published year of references	Short text	Unitless
Reference's DOI	Digital Object Identifier of references	Short text	Unitless

[a]The field only appears in ^{7}Be or ^{210}Pb annual concentration worksheet;

[b]The field only appears in ^{7}Be or ^{210}Pb annual depositional flux worksheet.

3 Results and discussions

3.1 Scope of the dataset

From 456 references (Appendix A), we have compiled a comprehensive dataset of atmospheric ^{7}Be and ^{210}Pb measurements made by numerous laboratories. The dataset includes 494 annual surface air concentration data of ^{7}Be covering 367 different sites, 366 annual surface air concentration data of ^{210}Pb from 270 different sites, 304 annual depositional flux data of ^{7}Be from 279 different sites, and 645 annual depositional flux data of ^{210}Pb from 602 different sites. In some cases, data collected from different periods were published in different articles. In these cases, all data from the same site are listed as separate dataset, but in data analysis and plotting, the data from the same site are merged. The sampling locations of this global dataset show broad geographical coverage (Fig. 1). The sampling maps are mainly composed of continental sites, while the oceanic monitoring sites are limited.

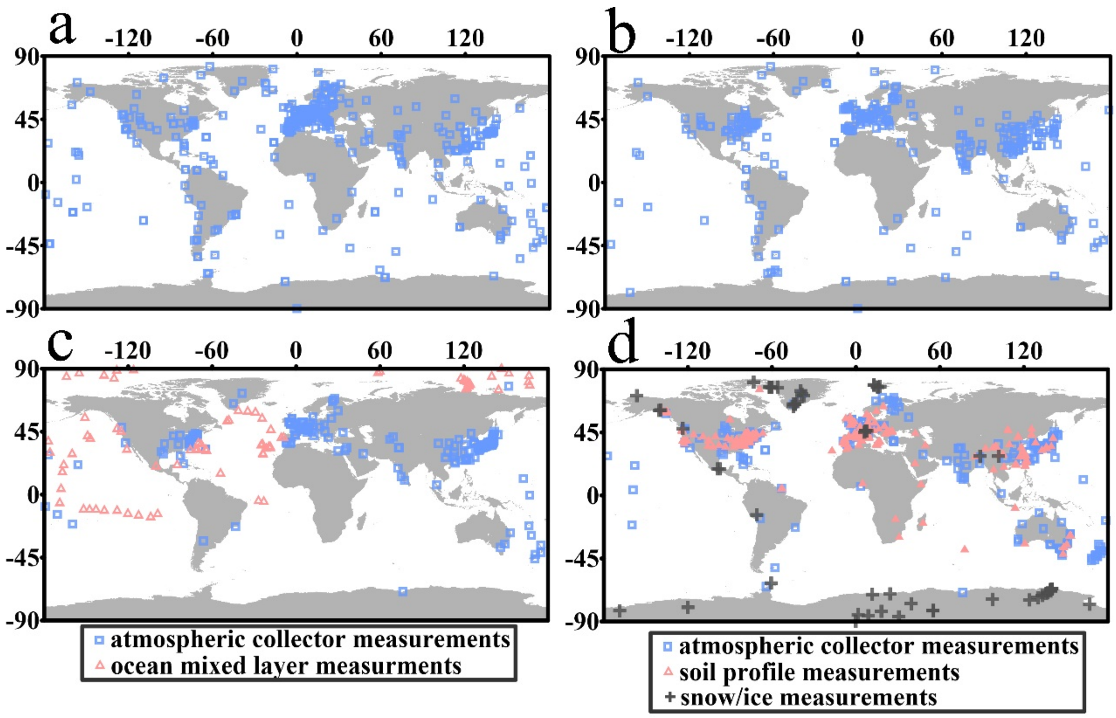

Figure 1: Maps showing sampling locations of (a) ^{7}Be concentration in surface air, (b) ^{210}Pb concentration in surface air, (c) ^{7}Be atmospheric depositional flux, and (d) ^{210}Pb atmospheric depositional flux.

The number of peer-reviewed journal articles published annually, containing ^{7}Be and ^{210}Pb data from 1955 to 2020 which are included in this article is plotted in Fig. 2. Measurements of ^{7}Be began in the mid-1950s (Arnold and Al-Salih, 1955; Cruikshank et al., 1956; Rama and Zutshi, 1958), earlier than those of ^{210}Pb, which began in early 1960s (Burton and Stewart, 1960; Crozaz et al., 1964; Peirson et al., 1966). The long-term monitoring work started in the 1980s with the ^{7}Be and ^{210}Pb concentration data generated by the EML Surface Air Sampling Program (Feely et al., 1989; Larsen et al., 1995). This was followed by more ambitious international programs such as the REM network (Hernandez-Ceballos et al., 2015) and IMS-CTBTO (Terzi and Kalinowski, 2017). However, in these two programs, ^{210}Pb concentration measurements were conducted only in a few stations (Heinrich et al., 2007; Sangiorgi et al., 2019). In contrast, direct ^{7}Be and ^{210}Pb flux measurements were rarely supported by the international program, but there were several national monitoring programs initiated by developed countries like Australia (Bonnyman and Molina-Ramos, 1976), Japan (Narazaki et al., 2003; Yamamoto et al., 2006), United States (Lamborg et al., 2013) and Finland (Paatero et al., 2015; Leppanen, 2019). The measurement of ^{7}Be inventory in ocean mixed layer began in the 1970s (Silker, 1972; Young and Silker, 1974, 1980), and the idea proposed by Young and Silker was subsequently developed in the upper 100-200 m to assess surface water subduction, oxygen utilization and rate of upwelling (Kadko, 2009; Kadko and Olson, 1996; Kadko and Johns, 2011). The measurement of $^{210}Pb_{ex}$ in an undisturbed soil profile was first conducted by Fisenne (1968). Subsequently, Benninger et al. (1975) and Moore and Poet (1976) showed that excess ^{210}Pb activities in undisturbed soil profiles can be utilized to estimate the atmospheric ^{210}Pb depositional flux, which resulted in an increase in the measurements of $^{210}Pb_{ex}$ in soil profiles in the late 1980s (Graustein and Turekian, 1986, 1989; Monaghan et al., 1989; Dörr and Munnich, 1991). Subsequently, $^{210}Pb_{ex}$ was shown to be useful for soil erosion studies on agricultural land (Walling and He, 1999; Walling et al, 2003).

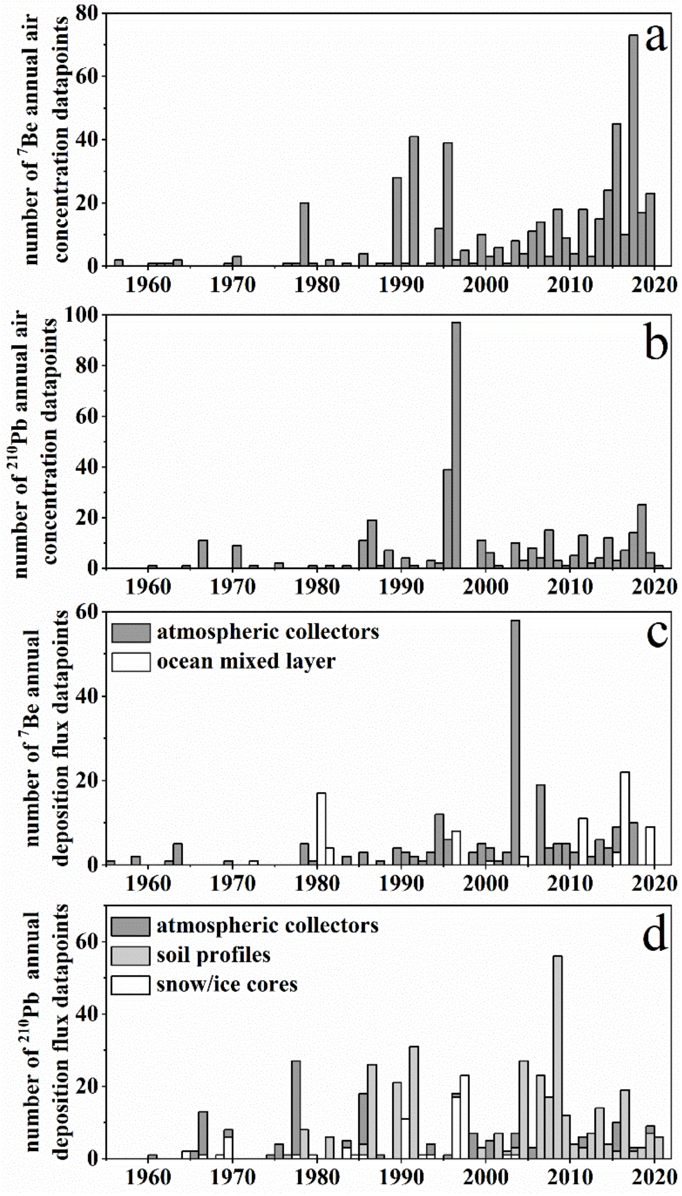

Figure 2: Distributions of data published between 1955 and early 2020: (a) ^{7}Be concentration, (b)^{210}Pb concentration, (c) ^{7}Be depositional flux, and (d) ^{210}Pb depositional flux.

The histogram of sampling durations of ^{7}Be and ^{210}Pb measurements is given in Fig. 3. In general, the duration of sampling for ^{7}Be measurements, especially for air concentration, is longer than that of ^{210}Pb. Globally, there are 140 sites that monitored ^{7}Be air concentration for more than 10 years. The long-term (decades) measurements of ^{7}Be were mainly dedicated to investigate the effect of changes in sunspot number on ^{7}Be (Megumi et al., 2000; Cannizzaro et al., 2004; Kulan et al., 2006; Pham et al., 2013; Steinmann et al., 2013). Due to the simpler measurement procedure for air concentration, the duration of air concentration measurements, whether for ^{7}Be or ^{210}Pb, is generally longer than that of ^{210}Pb and/or ^{7}Be depositional flux measurements.

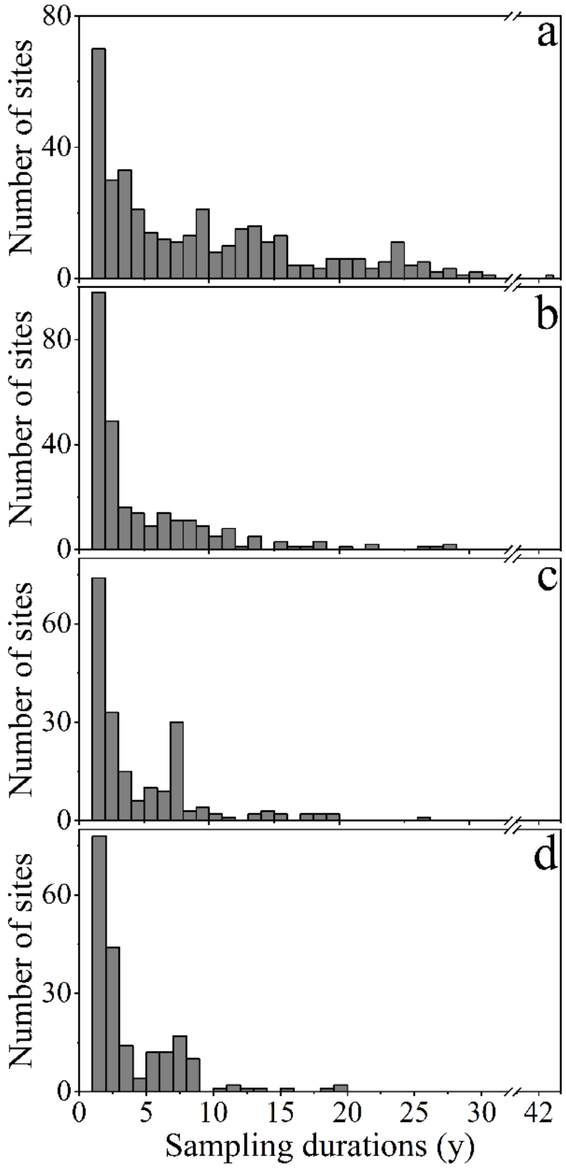

Figure 3: Histogram of sampling durations of (a) ^{7}Be concentration, (b) ^{210}Pb concentration, (c) ^{7}Be depositional flux, and (d) ^{210}Pb depositional flux. For those sites with multiple dataset, we added the sampling duration together, after deducting the overlapping period, if any. Note that the ^{7}Be and ^{210}Pb depositional flux data plotted here refer to those obtained using rain collectors.

3.2 Global variability

The global data of ^{7}Be and ^{210}Pb air concentrations and depositional fluxes are presented in Fig. 4. The range of concentrations of ^{7}Be and ^{210}Pb are 0.33-17.77 mBq m^{-3} and 0.003-4.65 mBq m^{-3}, respectively. The range of depositional fluxes of ^{7}Be and ^{210}Pb are 59-6350 Bq m^{-2} y^{-1} and 1-2539 Bq m^{-2} y^{-1}, respectively. The concentrations and depositional fluxes of ^{7}Be show discernable latitudinal variability (Fig. 5a and 5c). In general, ^{7}Be concentration and flux peak at the mid-latitudes and decrease toward the equator and poles, as was theoretically predicted by Lal and Peters (1967). A symmetric pattern is observed between the Northern and Southern hemispheres, however, a sharp increase in ^{7}Be air concentration (lack of flux data) occurred on the Antarctic, which reflects the subsidence of stratospheric air masses over the Antarctica continent (Wagenbach et al., 1988; Elsässer et al., 2011). Although the ^{210}Pb concentration and depositional flux are expected to heavily depend on the source(s) of air mass(es), and not to depend on the latitude, the 10° latitudinal variability of ^{210}Pb concentration and depositional flux in the Northern and Southern hemisphere is observed (Fig. 5b and 5d). The latitudinal variability of ^{210}Pb flux is similar to the global fallout curve based on 167 global sites (Baskaran, 2011). Since most of the ^{210}Pb data are derived from continental sites, the latitudinal variation is mostly due to differences in the radon emanation rates with latitude. As the area of the landmass in the Southern hemisphere is smaller compared to the Northern hemisphere, the ^{210}Pb concentration and depositional flux are much lower there. An asymmetry is observed in the ^{210}Pb concentration and depositional flux between the Northern and Southern hemispheres, with the highest values appearing in the mid-latitudes of the Northern hemisphere.

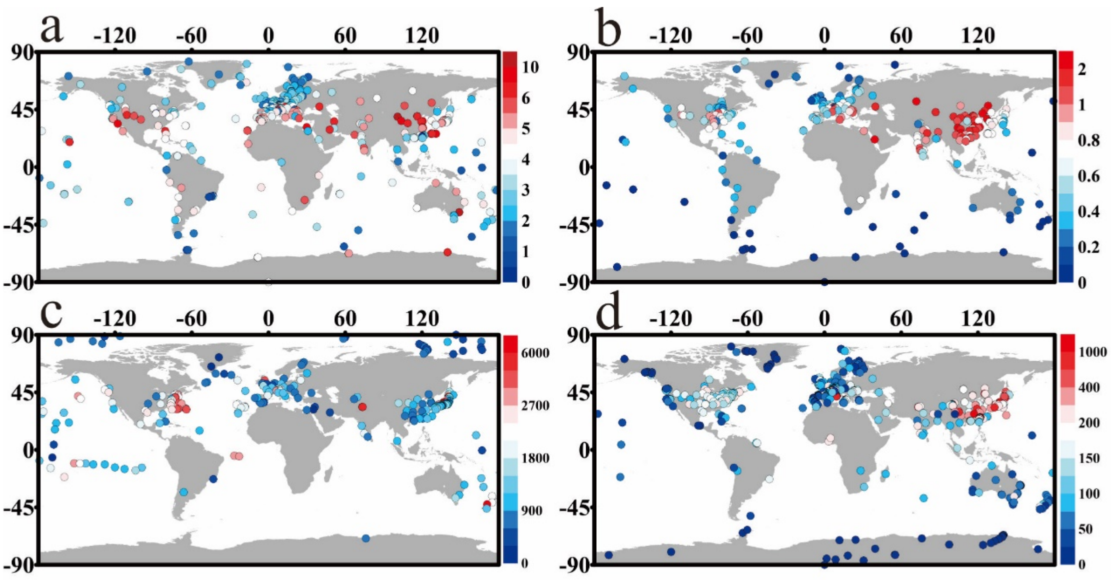

Figure 4: Global distribution of (a) ^{7}Be annual air concentration (mBq m^{-3}), (b) ^{210}Pb annual air concentration

(mBq m^{-3}), (c) ^{7}Be annual depositional flux (Bq m^{-2} y^{-1}) and (d) ^{210}Pb annual depositional flux (Bq m^{-2} y^{-1}). For those sites with multiple dataset, a weighted average was calculated based on sampling durations.

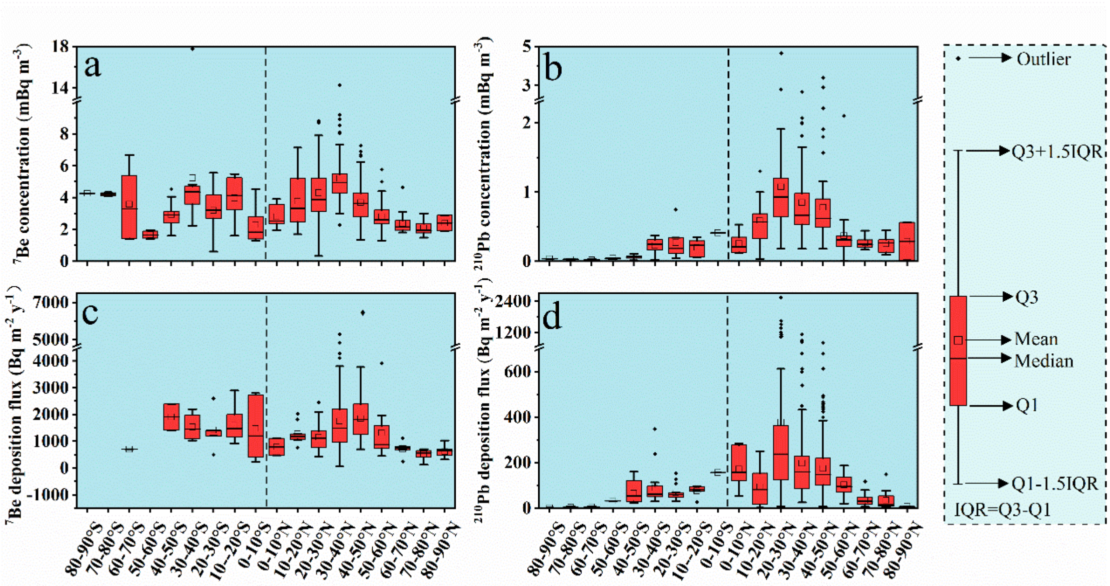

Figure 5: Latitudinal variability (box whisker plots) of (a) ^{7}Be concentration, (b) ^{210}Pb concentration, (c) ^{7}Be depositional flux, and (d) ^{210}Pb depositional flux plotted by 10° latitudinal bands.

3.3 Fraction of dry deposition and effect of precipitation on ^{7}Be and ^{210}Pb depositional flux

It was reported that dry deposition of ^{7}Be and ^{210}Pb generally accounts for less than 10% of the total deposition (Talbot and Andren, 1983; Brown et al., 1989; Todd et al., 1989), however, the fraction of dry deposition of ^{7}Be and ^{210}Pb is highly variable (McNeary and Baskaran, 2003; Pham et al., 2013). It is likely that the contribution of dry fallout could increase when annual precipitation decreases (McNeary and Baskaran, 2003). The fraction of dry to total depositional flux of ^{7}Be and ^{210}Pb are presented in Fig. 6a and 6b. Globally, the fraction of dry to total depositional flux of ^{7}Be and ^{210}Pb ranged from 1% to 44% (mean:12±9%, n=29, excluding one extreme site without precipitation) and from 5% to 51% (mean: 21±12%, n=26), respectively (Fig. 6c). The low fraction of dry to total depositional fluxes of ^{7}Be and ^{210}Pb suggest that these nuclides are removed from the atmosphere primarily by precipitation (both rain and snowfall). Our results also support previous studies (Baskaran et al., 1993; Benitez-Nelson and Buesseler, 1999) that the fraction of dry deposition is higher for ^{210}Pb than for ^{7}Be. The fraction of dry fallout of ^{7}Be and ^{210}Pb is plotted against annual precipitation in Fig. 6d and 6e, a weak negative correlation is observed especially for ^{210}Pb.

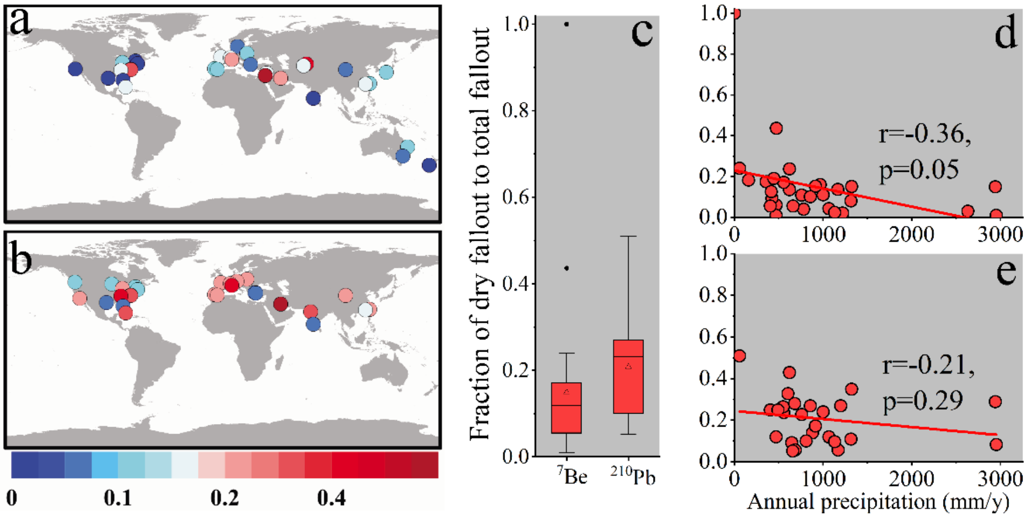

Figure 6: Global distribution of the fraction of dry to total depositional fluxes of (a)^{7}Be and (b) ^{210}Pb. (c) Box whisker plots showing the comparison between the fraction of dry to total depositional fluxes of ^{7}Be and the fraction of dry to total depositional fluxes of ^{210}Pb. (d) The fraction of dry to total depositional fluxes of ^{7}Be versus annual precipitation. (e) The fraction of dry to total depositional fluxes of ^{210}Pb versus annual precipitation.

As precipitation is the primary mechanism of removal of these nuclides from the atmosphere, the annual depositional fluxes generally depend on the amount and frequency of precipitation. In our dataset, the world's lowest ^{7}Be depositional flux (only 59 Bq m^{-2} y^{-1}, less than 5% of the global average ^{7}Be flux) occurred in Judean Desert, a precipitation free area in the horse latitude (Belmaker et al., 2011). The highest ^{7}Be (6350 Bq m^{-2} y^{-1}) and ^{210}Pb (2539 Bq m^{-2} y^{-1}) depositional flux were observed in heavy rainfall areas, Hokitika, New Zealand (Harvey and Matthews, 1989) and Taiwan (Huh and Su, 2004), respectively. Positive correlations between annual depositional flux and precipitation have been observed on a local scale (e.g., Narazaki et al., 2003; Garcia-Orellana et al., 2006; Sanchez-Cabeza et al, 2007; Leppanen et al., 2019). Here we illustrate the effect of precipitation on annual depositional flux on a global scale (Figs. 7 and 8). As both ^{7}Be and ^{210}Pb depositional flux show latitudinal variability, the linear best-fit curve of annual depositional flux to annual precipitation is plotted within the 10° latitudinal bands (if data are available). The empirical equations and fitting parameters describing the relationships between annual precipitation and ^{7}Be and ^{210}Pb depositional fluxes are summarized in Table 2. ^{7}Be and ^{210}Pb annual depositional fluxes generally show a good positive correlation with annual precipitation, although the data are limited in some latitudinal bands. Note that the frequency of precipitation is also an important factor which is not considered in earlier studies.

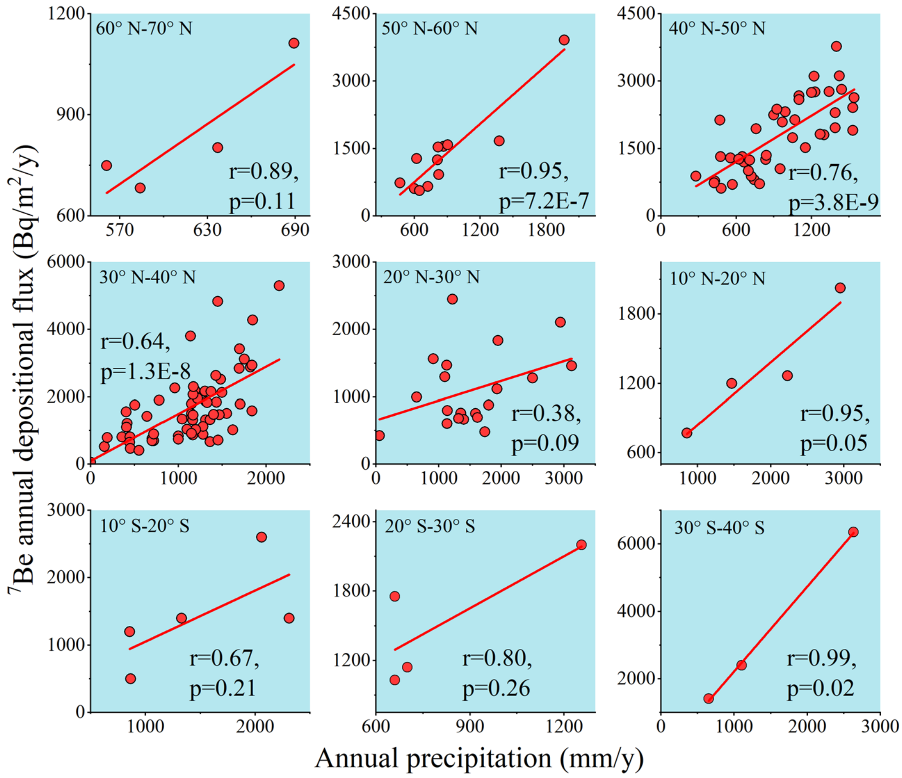

Figure 7: Annual depositional fluxes of ^{7}Be are plotted against annual precipitation for 10° latitudinal bands.

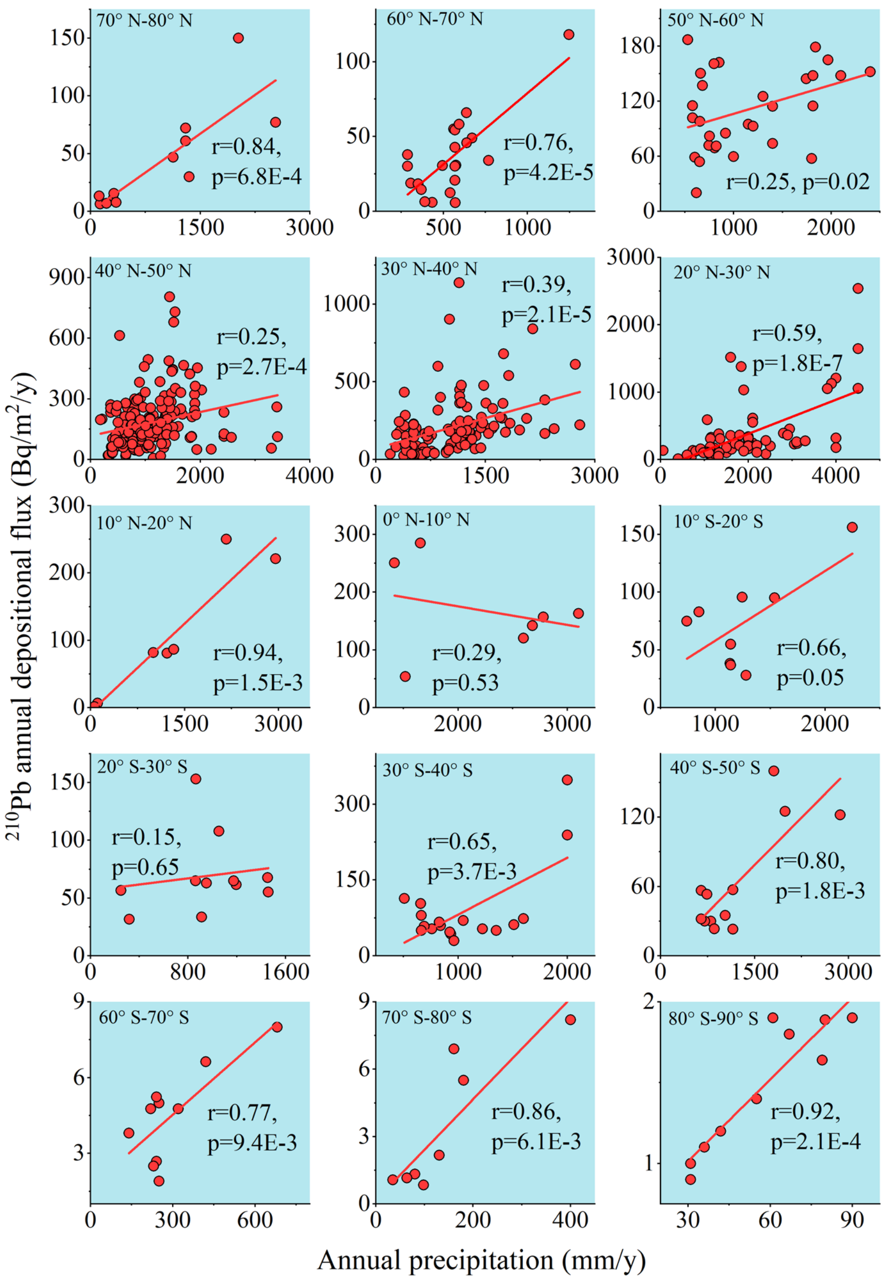

Figure 8: Annual depositional fluxes of ^{210}Pb are plotted against annual precipitation for 10° latitudinal bands.

3.4 $^{7}Be/^{210}Pb$ ratios and deposition velocities

Considering that some data come from the same station, we further calculated the ratios of ^{7}Be to ^{210}Pb and deposition velocities of aerosols using ^{7}Be and ^{210}Pb data, as shown in Figs. 9 and 10.

The variations in the $^{7}Be/^{210}Pb$ ratios reflect both vertical and horizontal transport in the atmosphere (Baskaran, 1995; Koch et al., 1996; Arimoto et al., 1999; Lee et al., 2007; Tositti et al., 2014). Our dataset exhibits similar global patterns of $^{7}Be/^{210}Pb$ ratio as simulated with a three-dimensional chemical tracer model (Koch et al., 1996), with a positive south poleward gradient and a little variation in the Northern hemisphere. Globally, the $^{7}Be/^{210}Pb$ air concentration ratio ranged from 2 to 222, and the $^{7}Be/^{210}Pb$ depositional flux ratio ranged from 2 to 229. In 19 sites for which $^{7}Be/^{210}Pb$ air concentration ratio and depositional flux ratio were available simultaneously, a paired t-test indicates that at 0.05 level the $^{7}Be/^{210}Pb$ air concentration ratio and depositional flux ratio are not significantly different.

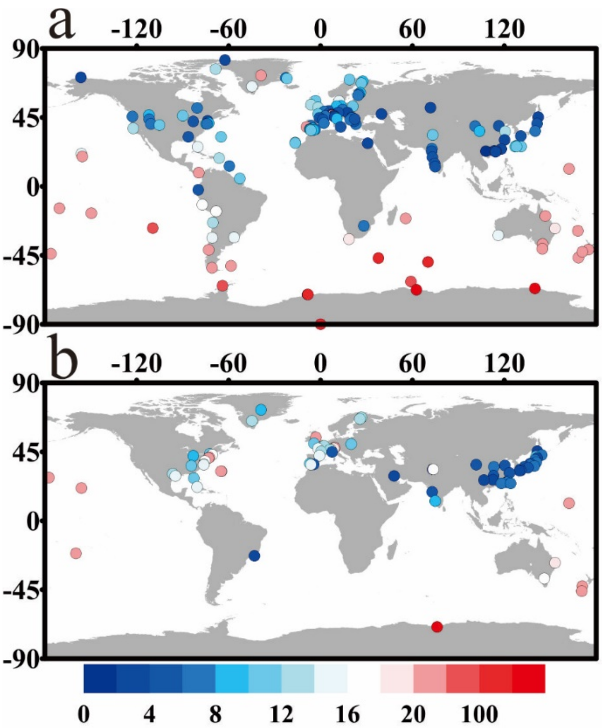

Figure 9: Global distribution of (a) $^{7}Be/^{210}Pb$ activity ratio and (b) $^{7}Be/^{210}Pb$ depositional flux ratio. Note that the color bar below applies to both figures.

^{7}Be and ^{210}Pb are excellent tracers for the determination of the deposition velocities of aerosols for two reasons: (1) their depositional fluxes and air concentrations at any given site remain fairly constant over a long period and (2) their size distributions in aerosols are similar to that of many particulate contaminants of interest (McNeary and Baskaran, 2003; Dueñas et al., 2005). When both air concentration (C) and depositional flux (F) at the same site are available, the average total deposition velocities of aerosols that carry these nuclides (V_d) can be calculated by the following Eq. (1):

$$V_d = F/C, \quad (1)$$

Thus, the V_d obtained from ^{7}Be and ^{210}Pb can be used to determine the depositional flux of analog species with a knowledge of their air concentration (Turekian et al., 1983). The V_d for ^{7}Be ranged from 0.2-8.4 (mean: 1.3±1.2, n=70) cm s^{-1}, and for ^{210}Pb ranged from 0.1-12.7 (mean: 1.2±1.7, n=72) cm s^{-1}. The deposition velocity of aerosols collected over a period of 17 months in Detroit, MI, USA varied over two orders of magnitude, from 0.2 to 3.6 (mean: 1.6, n=30) cm s^{-1} for ^{7}Be and 0.04 to 3.6 cm s^{-1} (mean: 1.1, n=30) cm s^{-1} (McNeary and Baskaran, 2003). A summary of deposition velocity from 10 different stations are also given in McNeary and Baskaran (2003). Earlier studies suggested that, at continental sites, V_d of ^{7}Be will be higher than V_d of ^{210}Pb using the ground level as the reference, which is an artifact in the manner in which the calculation is made (Turekian et al., 1983; Todd et al., 1989; McNeary and Baskaran, 2003). However, later works observed opposite results (Dueñas et al., 2005, 2017; Lozano et al., 2011; Mohan et al., 2019). The independent t-test analysis indicates that at 0.05 level the V_d calculated by ^{7}Be and ^{210}Pb are not significantly different in the global dataset, which suggests that ^{7}Be and ^{210}Pb attach onto the aerosols by similar mechanisms (Winkler et al., 1998; Papastefanou, 2006), and are affected by similar deposition processes (Lozano et al., 2013).

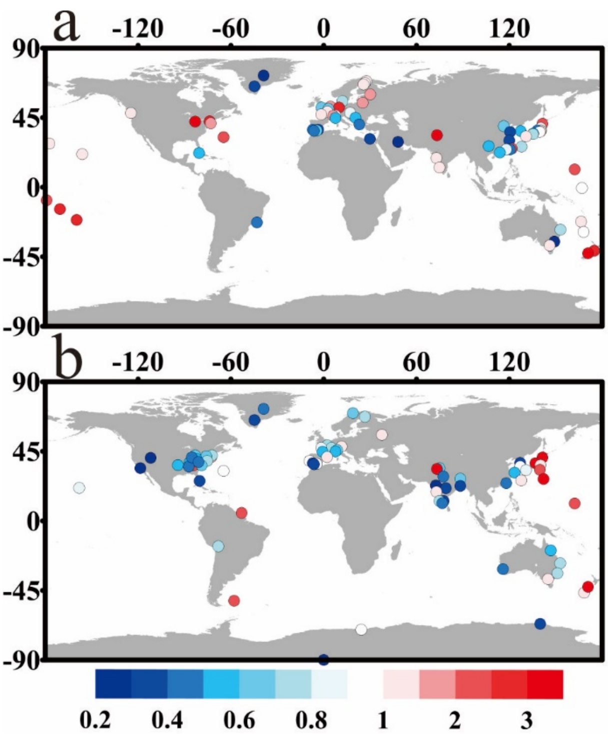

Figure 10: Global distribution of deposition velocities (V_d) of aerosols (in cm s^{-1}) obtained from (a)^{7}Be and (b) ^{210}Pb. Note that the color bar below applies to both figures.

3.5 Investigations on global atmospheric dynamics and climate changes

Developing numerical models in which aerosols, chemistry, radiation, and clouds interact with one another and with atmospheric dynamics is important for understanding and predicting global climate changes (Brost et al., 1991). In such atmospheric dynamic models, the major uncertainty is from the parameterization of subgrid-scale processes such as precipitation scavenging, vertical transport, and radiative effect. The cosmogenic ^{7}Be and terrigenous ^{210}Pb, taken together, offer an excellent tool in investigating wet scavenging and vertical transport in global models (Liu et al., 2001).

A set of data obtained prior to the 1990s was used to compare simulated results in global models such as ECHAM2 (Brost et al., 1991; Feichter et al, 1991), ECMWF (Rehfeld and Heimann, 1995), CTM based on GISS GCM (Balkanski et al., 1993; Koch et al., 1996), GEOS-CHEM (Liu et al., 2001), LMDz (Preiss and Genthon, 1997; Heinrich and Pilon, 2013) and GMI CTM (Liu et al., 2016). By simulating the ratio of $^{7}Be/^{210}Pb$, Koch et al. (1996) eliminated the error associated with the effect of precipitation and provided a better measure of vertical transport. After correcting the cross-tropopause transport, simulation of observed ^{7}Be and ^{210}Pb surface concentrations and depositional fluxes with no significant global bias was obtained (Liu et al., 2001). Note that the spatial coverage of the dataset used in the previous modeling work was only partial, thus, limiting the statistical significance of comparisons of simulated and observed results (Feither et al., 1991). Additional work with more data is needed for detailed comparison and successful validation of models (Brost et al., 1991). The size of the ^{7}Be and ^{210}Pb datasets has greatly increased in the last three decades and our new dataset is expected to lay a foundation in developing better parameterization and contribute to modeling efforts.

3.6 Soil erosion, aquatic particle dynamics and ocean surface process studies

The atmospheric depositional flux data of ^{7}Be and ^{210}Pb are useful in utilizing these nuclides as tracers for soil erosion and redistribution studies in terrestrial environments (Mabit et al., 2008) as well as particle dynamics study in aquatic environments (e.g., Du et al., 2012; Matisoff et al., 2014). The basic principles involved in using of ^{7}Be or $^{210}Pb_{ex}$ as soil tracers are the same, which is to compare the measured inventory of ^{7}Be or $^{210}Pb_{ex}$ (Bq m^{-2}) at a sampling point with the inventory at an undisturbed (or reference) site (Blake et al., 1999; Walling and He, 1999). Depletion of the inventory means that soil erosion has occurred, whereas enrichment provides evidence of accumulation of surficial soil. The first step in these studies is to select a suitable undisturbed site and obtain the reference inventory (Mabit et al., 2009). However, as human activity intensifies, such undisturbed sites are not always readily available. In aquatic systems (including river, lake, estuary and coast), the mass balance models of ^{7}Be and $^{210}Pb_{ex}$ have become powerful tools to understand the sediment source, transportation and resuspension processes (e.g., Wieland et al., 1991; Feng et al., 1999; Jweda et al., 2008; Huang et al., 2013; Mudbidre et al., 2014). In such models, the atmospheric depositional input of ^{7}Be and ^{210}Pb is a required source term. Furthermore, $^{7}Be/^{210}Pb_{ex}$ activity ratio can be used to identify the source area of sediments (Whiting et al., 2005; Jweda et al., 2008; Wang et al., 2021), to quantify the age of sediments (Matisoff et al., 2005;

Saari et al., 2010), and to determine the transport distance of suspended particles (Bonniwell et al., 1999; Matisoff et al., 2002). Thus, the atmospheric depositional flux data of ^{7}Be and ^{210}Pb are also important for tracing particle dynamics in aquatic systems.

The atmospheric depositional flux (or ocean inventory) data of ^{7}Be serve as an indispensable parameter for tracing surface ocean process (e.g., subduction, upwelling, and depositional flux of trace metals) (Kadko, 2017; Kadko and Olsen, 1996; Kadko et al., 2015). Due to the low activity of ^{7}Be in open ocean waters, usually, 400-700 L seawater is needed, which imposes some limitations for sampling, especially for deep layers. This constraint has limited its application. If the ^{7}Be ocean inventory can be accurately estimated, the collection of a large volume of seawater can be avoided and the application of ^{7}Be in open ocean will be expanded.

Scientific data are not simply the outputs of research but they also provide inputs to new hypotheses, extending research and enabling new scientific insights (Tenopir et al., 2011). Our dataset provides a forum in which a large amount of ^{7}Be and ^{210}Pb atmospheric depositional flux data for the above-mentioned research communities. This database will help in identifying data gaps and evaluating the empirical relations between ^{7}Be and ^{210}Pb depositional fluxes and annual precipitation (Table 2). Researchers can rely on previously collected data in planning their research, without additional monitoring of ^{7}Be and/or ^{210}Pb depositional fluxes. Even for those areas with data gaps, the empirical equations between ^{7}Be and ^{210}Pb depositional fluxes and annual precipitation provide an empirical method for estimating fluxes, especially for ^{7}Be, as ^{7}Be depositional flux is independent of longitude and is constant over broad latitudinal bands. In summary, the atmospheric depositional flux data presented in this our dataset along with the meta-analysis of the data will be useful in the investigations of soil erosion studies in terrestrial environments, particle dynamics studies in aquatic systems, and surface mixing process studies in open ocean.

3.7 Gaps and recommendations

Our dataset is a compilation of most of the published results in international peer-reviewed journal articles. Although the spatial coverage of this dataset is significant, the data available sites are unevenly distributed. Compared to spatial and temporal coverage of depositional flux data, the coverage of air concentration data is much larger. The air concentration measurement from deep ocean site is limited, but the data from coastal oceanic sites is adequate; however, the depositional flux measurement at oceanic sites is rare. Concerning air concentrations, areas such as Europe, East Asia, eastern Oceania, and the eastern United States are well covered, whereas other areas such as the African continent and Northern Asia are limited. A similar spatial coverage pattern exists for the depositional flux of these nuclides, but the regional gaps are more notable, especially for ^{7}Be flux data which are almost non-existent in Antarctica and African continents. In addition, it needs to be emphasized that the number of sampling sites, in which both concentration and flux of ^{7}Be and ^{210}Pb were measured simultaneously, are limited.

We recommend that future studies should pay more attention to those areas that are currently undersampled or unsampled to better characterize the expected global variability in the ^{7}Be and ^{210}Pb air concentrations and depositional fluxes, by measuring both nuclides simultaneously to obtain more data as well as ^{7}Be/^{210}Pb ratio and estimate deposition velocity of aerosols. In areas with very limited precipitation such as deserts, it is expected that the dry fallout will dominate the bulk depositional flux, and quantification of the role of dry fallout in the removal of these nuclides will provide insights on the removal of other analog species. As mentioned earlier, combining cosmogenic ^{7}Be with ^{210}Pb that has a predominantly Earth-surface origin will be useful to trace species that originate both from Earth' surface, such as Hg, SO_4^{2-}, NO_3^-, and those that originate in the upper atmosphere, such as O_3. The size distribution of aerosols particles carrying ^{7}Be and ^{210}Pb is crucial for understanding atmospheric behavior and tracing analogues, and such studies also need to be conducted. Besides, the troposphere contains ~99% of global water vapor with < 1% in the stratosphere. The deposition velocity of aerosol in the stratosphere is very low (~ 4×10^{-3} cm s^{-1}; Junge, 1963) with no precipitation. Thus, the ^{7}Be concentration is governed by local production, zonal and vertical downward transport, and its decay. In the middle and upper troposphere where precipitation is much less frequent, the removal rate of aerosols is also slow. Collection of air samples in that part of the atmosphere will provide useful information on the total deposition velocity of aerosols (Lal and Baskaran, 2012).

Finally, we acknowledge that the seasonal data of ^{7}Be and ^{210}Pb has not been included in the current version of dataset, because compiling the seasonal data is more challenging than compiling the annual data. Unlike the annual data, most of the published seasonal data are presented in graphs, without giving in tables, and in some cases, the graph quality was poor and precision in data extraction is expected to be poor. Besides, in some papers, although seasonal data were measured, only the annual data were provided. Thus, the comprehensive compilation of seasonal data of ^{7}Be and ^{210}Pb may need collaboration with and data sharing from the scientific community. The compilation of seasonal data is expected to be useful to assess seasonal variability of ^{7}Be and ^{210}Pb and understand the relationship between these changes and influencing factors such as atmospheric dynamics, meteorological conditions, and geographic location on a global scale. And the seasonal data can also be useful in evaluating seasonality of transport in global atmospheric models.

4 Data formats and availability

For clarity and convenience, four separate worksheets, each named as ^{7}Be or ^{210}Pb annual air concentration and ^{7}Be or ^{210}Pb annual atmospheric flux, are available in one Microsoft Excel® file, although sometimes these data come from the same article. In addition, the data of $^{7}Be/^{210}Pb$ air concenration ratio, $^{7}Be/^{210}Pb$ depositional flux ratio and deposition velocities for ^{7}Be and ^{210}Pb are also presented. The dataset can be downloaded from Zenodo (https://doi.org/10.5281/zenodo.4785136, Zhang et al., 2021). It is free for scientific applications, but the free availability does not constitute a license to reproduce or publish it. The compilation of seasonal data is ongoing, and the dataset will be updated in a future effort.

5 Conclusions

This paper summarizes the global dataset of ^{7}Be and ^{210}Pb for their concentration in atmospheric air and their depositional fluxes from 456 publications spanning the period from 1955 to early 2020. The calculated activity ratios of $^{7}Be/^{210}Pb$ and deposition velocity of aerosols are also reported. Some noteworthy spatial gaps in the dataset are the African continent, Northern Asia, and Antarctica (only for ^{7}Be flux). Despite these gaps, our dataset is the largest compilation of ^{7}Be and ^{210}Pb air concentration and depositional flux up to date and could be used to better understand the transport processes of air masses and depositional processes of aerosols. This dataset not only lays a solid foundation to develop better parameterization contributing to future modeling efforts but also supply a basic parameter for tracing soil erosion on land, particle dynamics in aquatic systems, and ocean surface processes using ^{7}Be and/or ^{210}Pb.

Appendix A: List of references in the dataset

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Author contributions

FZ is responsible for most of the writing of this article, along with the assembly of the data and preparation of the figures. QZ and YW assisted FZ in compiling the data. JW, MB, QZ, PJ and JD contributed to the review of the manuscript.

Competing interests

The authors declare that they have no conflict of interest.

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