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Atmospheric ^{210}Pb, ^{210}Po and ^{210}Po/^{210}Pb activity ratio in urban aerosols: temporal variability and impact of biomass burning emission

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ABSTRACT

Atmospheric ^{210}Pb, ^{210}Po and their activity ratio (^{210}Po/^{210}Pb) have been studied for two years (January 2007–April 2009) from an urban site (Kanpur: 26.5°N and 80.3°E) in the Indo-Gangetic Plain. The average activities of ^{210}Pb and ^{210}Po centre on 1.8 mBq m^{-3} (range: 0.5–4.8 mBq m^{-3} for $n = 99$) and 0.094 mBq m^{-3} ($n = 21$, range: 0.002–0.28 mBq m^{-3}), respectively. The temporal variability in the activity of ^{210}Pb is significantly pronounced, with relatively high levels during October–November and December–February; a trend similar to that observed for the carbonaceous species. The high aerosol abundance coinciding with the biomass burning emissions (agricultural-waste burning) during October–November and stagnant boundary layer in the wintertime (December–February) is the dominant factor for the observed temporal trend. The preliminary data suggest that biomass burning emissions also contribute to the atmospheric ^{210}Po activity, as evident from the large variability in the ^{210}Po/^{210}Pb activity ratio (range: 0.02–0.23) at this urban site. These results have implications to the model-based activity levels of ^{210}Pb and ^{210}Po from in-situ decay of the parent nuclide (^{222}Rn) for given latitude.

Keywords: atmospheric ^{210}Pb, ^{210}Po activities, biomass burning emission, Indo-Gangetic Plain

1. Introduction

The environmental radionuclides (e.g. ^{7}Be, ^{137}Cs, ^{210}Pb, ^{210}Po and ^{222}Rn) of natural and anthropogenic origin serve as a robust tool for studying various atmospheric processes. Among their several applications, most relevant is the ability to serve as tracers of continental air masses, long-range transport of aerosols (Sarin et al., 1999; Hammer et al., 2007), stratosphere–troposphere exchange rates (Kritz et al., 1991; Liu et al., 2004) and to assess the residence time of atmospheric aerosols (Marley et al., 2000; Baskaran and Shaw, 2001; Papastefanou, 2006; Lozano et al., 2011). The ^{210}Pb ($t_{1/2} = 22.3$ yr) is a naturally occurring radionuclide injected into atmosphere from the decay of ^{222}Rn ($t_{1/2} = 3.8$ d) emanated from the Earth's surface. The ^{210}Po ($t_{1/2} = 138$ d) is also formed in the atmosphere from the decay of ^{210}Pb via short-lived ^{210}Bi ($t_{1/2} = 5$ d). But unlike ^{210}Pb, atmospheric ^{210}Po is also contributed from other sources such as volcanic eruption (Nho et al., 1997), coal combustion (Kim et al., 2005), as a volatile species from coastal waters and sea-spray aerosols (Kim et al., 2000) and biomass burning emissions (LeCloarec et al., 1995; Nho et al., 1996).

In general, residence time of atmospheric aerosols varies from few days to weeks. In this context, ^{210}Pb – ^{210}Po and ^{210}Pb – ^{210}Bi pairs serve as a potential tool due to their particle-reactive nature and similar removal processes from the atmosphere (Marley et al., 2000). The ^{210}Pb – ^{210}Po pair utilises the radioactive disequilibrium among these two nuclides for the estimation of residence time of atmospheric aerosols (Rangarajan and Eapen, 1990; Marley et al., 2000; Baskaran and Shaw, 2001; Baskaran, 2011; Lozano et al., 2011). However, the concept of residence time is strictly applicable only in case of their in-situ production in the atmosphere from decay of the parent nuclide (^{222}Rn). Their direct injection into atmosphere from other sources (such as volcanic eruption and coal combustion) violates the basic assumption of constant production rate in the atmosphere.

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Despite their extensive usefulness, data on ^{210}Pb and ^{210}Po in atmospheric aerosols is rather sparse from urban sites (Rangarajan, 1992; Rastogi and Sarin, 2008). In this study, we report on the atmospheric ^{210}Pb, ^{210}Po and their activity ratio measured from an urban site (Kanpur, 26.5°N, 80.3°N, ~145 m above sea level) in Northern India. The main objective is to assess the temporal variability and factors influencing the activities of ^{210}Pb and ^{210}Po and ^{210}Po/^{210}Pb ratio in ambient aerosols. This study also represents the first data-set on ^{210}Pb and ^{210}Po from an urban location influenced by the widespread biomass burning emissions (agricultural-waste burning) in the Indo-Gangetic Plain (IGP).

2. Methodology

2.1. Aerosol sampling and site description

The PM_{10} (particulate matter with aerodynamic diameter $\leq 10\,\mu m$) samples were collected for a period of one year (January 2007–February 2008) from an urban site (Kanpur: 26.5°N, 80.3°E, ~142 m asl; Fig. 1) located at the centre of the IGP (Northern India). All samples ($n = 57$) were collected using a high-volume sampler (flow rate: $1.0 \pm 0.1\,m^3\,min^{-1}$) on pre-combusted tissuquartz filters (PALLFLEX^{TM}, 2500 QAT-UP; size: $20.0 \times 25.0\,cm^2$) for about 8–10 hrs. A second set of PM_{10} samples ($n = 42$) were collected from the same site during October 2008–April 2009.

The mass concentration of PM_{10} ($\mu g\,m^{-3}$) and its chemical composition at the sampling site (Kanpur) is influenced by the regional emission sources (biomass burning, industrial units and fossil-fuel combustion) and long-range transport of mineral dust (Ram et al., 2010). The emission from biomass burning (agricultural-waste and use of wood-fuel for domestic heating) is dominant during October–November and in the wintertime (December–February); representing an important source of carbonaceous aerosols along the entire stretch of the IGP. Mineral aerosols from the Desert regions in western India and from the Middle East contribute significantly to the aerosol loading during the summer months (April–June). The details on the temporal variability in chemical composition of aerosols have been reported in an earlier publication (Ram et al., 2010).

2.2. Analysis of ^{210}Pb in aerosols

A quarter portion of the sample filter (area = $105\,cm^2$) was cut into small strips and sealed in a plastic vial (height = 40 mm and diameter = 20 mm). The vial was placed inside a low-background High-Purity Germanium well detector (HPGe, Canberra Model GCW2523) coupled to a Digital Spectrum Analyser (Canberra Model DSA 100). The energy scale (0.4 keV per channel) was calibrated with 1173 and 1332 keV γ-peaks from a standard (^{60}Co) source. The activities of ^{210}Pb in samples were assayed by using 46.5 keV photo-peak (γ-yield: 4.05%). Depending on the ^{210}Pb activities in aerosols, counting time varied from about 9000 to 360 000 sec. In general, samples were counted to accumulate a minimum of 1000 counts under 46.5 keV energy region (with a spread of 2 keV envelope). The absolute counting efficiency of HPGe well detector at 46.5 keV was ascertained based on the known activity of ^{210}Pb from a standard solution of ^{238}U, deposited onto quarter portion of a blank filter and sealed in a plastic vial (similar to the sample vial); thereby matching the sample matrix and geometry. The ^{210}Pb activity in ^{238}U standard solution was quantitatively assayed by alpha-spectrometry (via ^{210}Po in presence of a ^{209}Po spike). An empirical factor of 11.05 ± 0.16 was used (normalised for self-absorption and geometry) based on the observed count rate at 46.5 keV and absolute ^{210}Pb activity in the standard source. The measured activities of ^{210}Pb in samples were decay corrected to the sampling date. The overall error, arising due to the counting statistics and decay correction is within ±10% (for total counts exceeding 1000). A number of repeat measurements ($n = 12$) show a good agreement for ^{210}Pb activities in the range of 0.71 ± 0.06 to $3.7 \pm 0.18\,mBq\,m^{-3}$. In order to check on the contribution of ^{210}Pb supported by ^{226}Ra associated with the regional soils, γ-peaks in the energy regions of 295, 352 and 609 keV were assessed in all aerosol samples. The count rate in these energy regions was not statistically different compared to the blank run with the filters. Along with samples, blank filters (quarter of the filter packed into a plastic vial with same geometry) were also analysed for ^{210}Pb. The average ^{210}Pb activity in blank filters was ~0.006 cpm (counts per minute) compared to that recorded in aerosol samples (Av = 0.6 cpm), and the contribution from blank filters to ^{210}Pb in samples was insignificant (~1%).

2.3. Analysis of ^{210}Po in aerosols

For the determination of ^{210}Po activity, a quarter of the sample filter was acid digested (with HF-HCl in presence of ^{209}Po spike) in a Teflon beaker. The clear solution was taken to near dryness followed by the addition of 2 ml of 6N HCl acid and further diluted to ~0.6N HCl with Milli-Q water (Resistivity = 18.2 MΩ-cm). To this final solution (~80 ml), ~500–600 mg of ascorbic acid was added to avoid interference from ferric ions (Fe^{3+}) before auto-plating Po isotopes (^{210}Po and ^{209}Po) onto Ag disc. The activities of ^{210}Po ($E_\alpha = 5.33\,MeV$) and ^{209}Po ($E_\alpha = 4.9\,MeV$) were assayed on silicon-surface-barrier

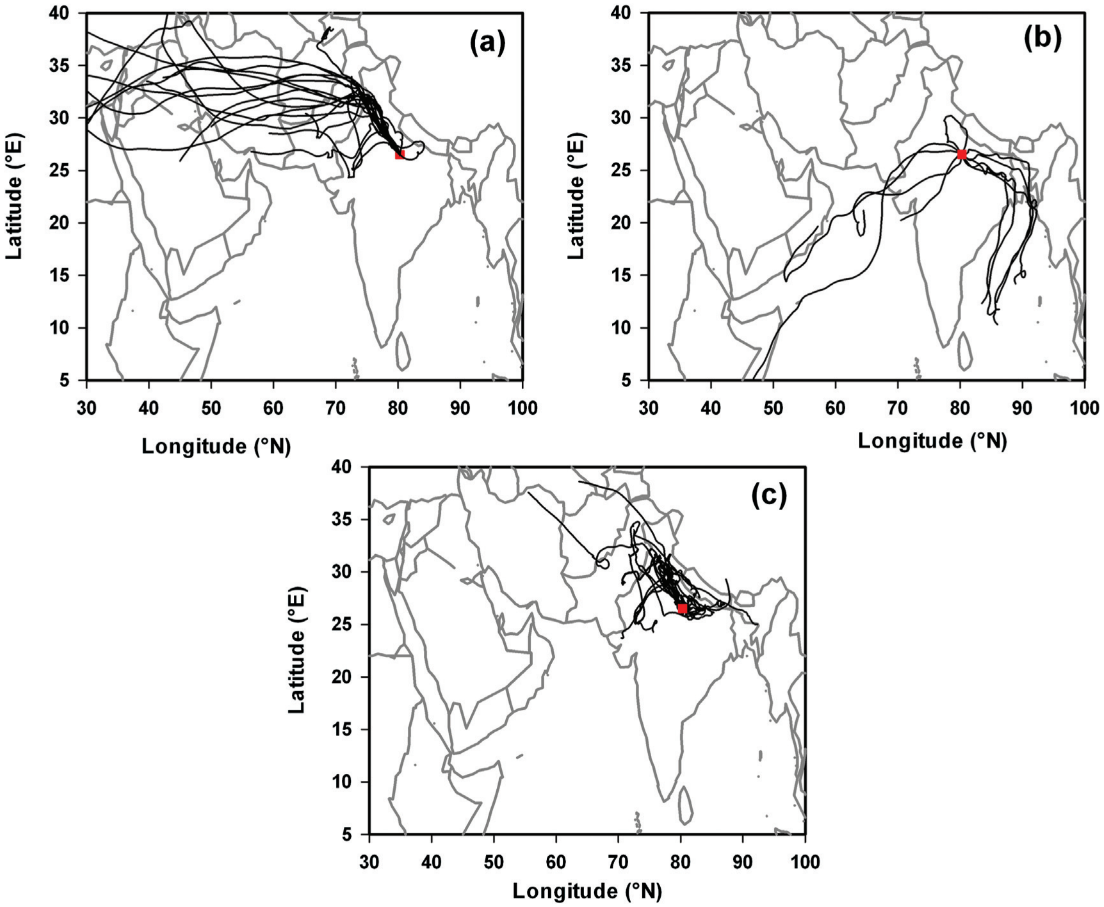

Fig. 1. Air mass back-trajectories ($n = 50$) arriving at the sampling site (Kanpur, shown in red-square) originating from (a) long-range transport from Desert regions (40%), (b) maritime air masses (22%) and (c) localised winds from Northern India (38%).

detectors (ORTEC Model 4001C) coupled to a multi-channel analyser (Canberra Series 35 Plus) (Sarin et al., 1999). The correction for decay of ^{210}Po from the time of plating to mid-counting; decay of ^{210}Po and its ingrowth from ^{210}Pb between the time of collection to time of their separation (plating date) were suitably incorporated for obtaining ^{210}Po activity in the aerosol samples. Blank filters were also analysed for ^{210}Po activity and was subtracted from the sample. The average ^{210}Po activity in blank filters was 0.15 ± 0.004 dpm (disintegrations per minute) compared to that in aerosol samples (Av: ~15.0 dpm). The ^{210}Pb activity in few of the aerosol samples ($n = 30$) was also measured by alpha-spectrometry. The ^{210}Pb activity measured by alpha spectrometry exhibit a close agreement with that assessed by gamma spectrometry (slope = 0.98, $R^2 = 0.66$, $n = 30$).

2.4. *Back-trajectory analysis*

The seven-day back-trajectory analysis was performed (at 500 m above ground level during October–February and 1000 m during March–June) to trace air masses arriving at Kanpur (Fig. 1). The air mass trajectories were computed from the final run data archive of Global Data Assimilation System (GDAS) model using National Oceanic and Atmospheric Administration (NOAA) Air Resource Laboratory Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (http://www.arl.noaa.gov/HYSPLIT) (Draxler and Rolph, 2003; Rolph, 2010). The back trajectories are computed for each one week during the entire sampling period, and a total of 50 back trajectories are computed during 2007–2009. The back trajectories are presented in Fig. 1 and are grouped into three categories, namely long-range transport from the Desert areas ($n = 20$, 40% of the total air masses; Fig. 1a), maritime ($n = 11$, equivalent to 22%; Fig. 1b) and localised over the North India ($n = 19$, 38% of the total; Fig. 1c). The long-range transport of air masses from Desert areas occurs during the late February–April and occasionally in January; whereas maritime air masses are characteristic of April–June. In contrast, the localised air masses are confined during October–November and in the wintertime.

It is important to state that the aerosol sampling was not considered to be of relevance during the period of southwest monsoon (July–September).

3. Results and discussion

3.1. Temporal variability of ^{210}Pb in ambient aerosols at Kanpur

The ^{210}Pb activity in PM$_{10}$ varied from 0.50 ± 0.02 to 4.8 ± 0.17 mBq m^{-3} (Fig. 2, Av: 1.8 ± 1.1 mBq m^{-3}; $n = 99$). The average ^{210}Pb activity at the sampling site (this study) is compared with other locations and summarised in Table 1. The ^{210}Pb activity in ambient aerosols at Kanpur is relatively high compared to the reported data in the literature (typically in the range of 1.0–2.0 mBq m^{-3}) (McNeary and Baskaran, 2003; Hammer et al., 2007; Rastogi and Sarin, 2008; May et al., 2009). The ^{210}Pb activity, averaged for each month during the entire sampling period, is presented in Fig. 3. The ^{210}Pb activity is significantly high during October–January and relatively low during March–June. The maximum activity of ^{210}Pb, 4.8 ± 0.17 mBq m^{-3} was observed in the samples collected during October–November.

The average ^{210}Pb activity at Kanpur exhibits a strong seasonal variability, nearly 2–3 times higher activity in October–November and December–February compared to that in April–June (Figs. 2 and 3). The seasonal variability in the ^{210}Pb activity can be explained in terms of ^{226}Ra content of regional soils, changes in the wind regimes and meteorological parameters (temperature, wind speed and boundary layer height). A remarkable similarity in the temporal variability for the abundance pattern of organic carbon (OC) and elemental carbon (EC) and ^{210}Pb activity provides strong evidence that biomass burning emissions and boundary layer dynamics in the IGP are the dominant factors for the observed data (Fig. 2). It has been proposed that the uptake of radioactive species (including ^{226}Ra, ^{238}U, ^{210}Po and ^{210}Pb) by food crops and plants (Vasconcellos et al., 1987; Manigandan, 2009; Rego, 2011) and direct injection of ^{210}Pb and ^{210}Po from biomass burning emissions could contribute significantly to the atmosphere. We suggest that large-scale post-harvest biomass burning emissions could serve as an additional source of ^{210}Pb and ^{210}Po, during October–February, over the urban atmosphere in the Gangetic Plain (see further discussion in Section 3.3).

The ^{226}Ra content of the regional soils also merits some discussion because of the emanation of the daughter nuclide ^{222}Rn ($t_{1/2} = 3.8$ d) which subsequently decays to ^{210}Pb. Studies on soil samples from different lithological units have suggested that the distribution of radionuclides

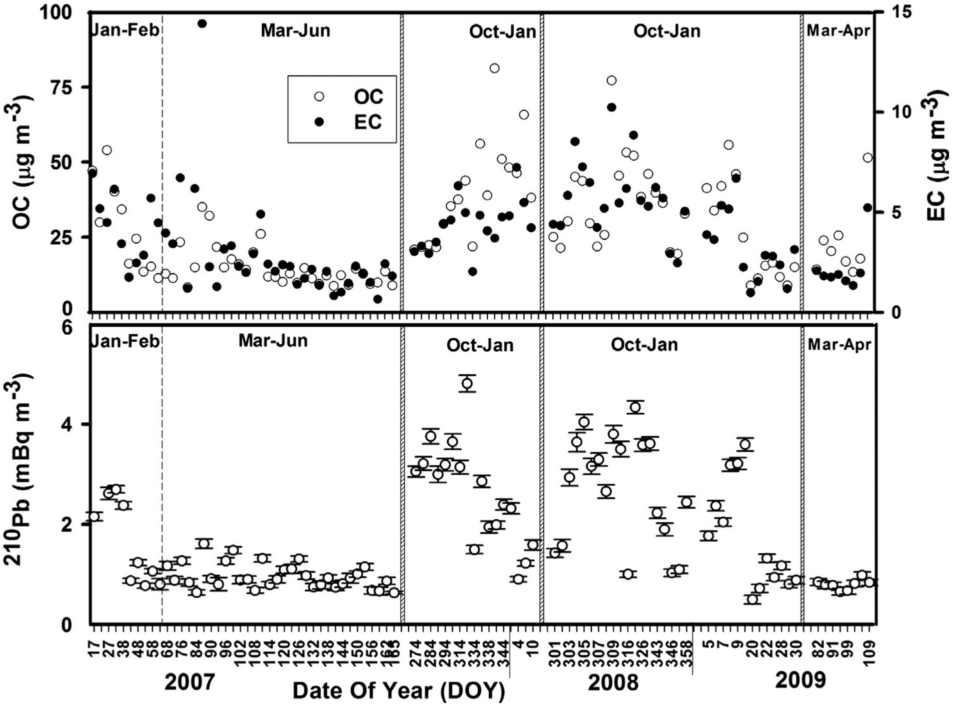

Fig. 2. Temporal variability of ^{210}Pb activity and carbonaceous species (EC, OC) in aerosols from Kanpur during the sampling period. The vertical dashed-line separates the seasons whereas the vertical bar indicates a break in the sampling.

depends on rock formation, their chemical properties and sample origin (Malakhov et al., 1966; Rao et al., 1996; Levin et al., 2002; Ramola et al., 2008; Baskaran, 2011). Rani and Singh (2005) had reported that ^{226}Ra concentration varied from 42.1 to 79.6 Bq kg^{-1} (Av: 57.3 Bq kg^{-1}) in soil samples collected from nearby areas in Himachal Pradesh. A soil sample collected from a nearby location from Kanpur and analysed for ^{226}Ra concentration shows significantly high level (182.6 Bq kg^{-1}) compared to that in soils from other parts of Northern India. Another study conducted from Hamirpur (31.7°N, 76.5°E) and Rajasthan (26.7°N, 73.5°E) suggests lower ^{226}Ra concentration in soils; the average ^{226}Ra concentration in soils from Hamirpur is reported to be 44.2 Bq kg^{-1} and it ranges from 30 to 70 Bq kg^{-1} in Rajasthan (Singh et al., 2003). The entire stretch of the IGP is dominated by the intense agricultural activity and the use of fertilizers (urea: NH_2CONH_2; $Ca_3(PO4)_2$ and $ZnSO_4/PO_4$) are very common. These fertilizers can concentrate ^{238}U and its daughter nuclides in the phosphate fraction. For example, phosphate rock samples and production of phosphate fertilizers, in Florida and Brazil, have reported high concentrations of ^{226}Ra, ^{228}Ra and ^{210}Pb radionuclides (Menzel, 1968; Fukuma et al., 2000). The indoor measurements of ^{222}Rn indicate higher concentrations (76 – 146 Bq m^{-3}) in some parts of Northern India (Singh et al., 2005). However, based on the assay of ^{226}Ra content in aerosol samples (as described in Section 2.2); we rule out any significant contribution of ^{210}Pb from ^{226}Ra via resuspension of regional soils.

The wind-rose plots for post-monsoon (October–November), wintertime (December–February) and summer months (April–June) during the course of this study are shown in Fig. 4. The average wind speed during winter and October–November is about two to three times lower than that during April–June (Ram et al., 2010). The dependence of ^{210}Pb activity on ambient temperature and wind speed is shown in Fig. 5. It can be seen that ^{210}Pb activity is relatively high during October–November and December–February, with the persisting lower ambient temperature and wind-speed conditions. These conditions in the wintertime are sustained by the shallow boundary layer height (500–900 m) along the Gangetic Plain (Nair et al., 2007). The ^{222}Rn activity in the ambient air has been used for estimating the planetary boundary layer dynamics and particularly the mixing height of regional pollutants (Malakhov et al., 1966; Levin et al., 2002; Zahorowski et al., 2005). This approach is usually adopted in view of the fact that the escape rate of ^{222}Rn is nearly constant over the land areas (~1 atom cm^{-2} s^{-1}) (Wilkening and Clements, 1975; Turekian and Graustein, 2003; Baskaran, 2011). A study reported in the literature has shown that the indoor ^{222}Rn activity levels exhibit

Table 1. ^{210}Pb, ^{210}Po and ^{210}Po/^{210}Pb activity ratios from an urban site (this study) are compared with the literature data. The values in the parenthesis represent average values.

Sites	Sampling period	*n*	Location	Elevation (m asl$^{\$}$)	^{210}Pb (mBq m^{-3})	^{210}Po (mBq m^{-3})	^{210}Po/^{210}Pb ratio	References
Kanpur	January 2007–April 2009	99	(26.5°N, 80.3°E)	142	1.8 ± 1.1	0.0–0.28 (0.094)	0.002–0.229 (0.079)	This study
Lodz, Poland	2008–2009		(51.8°N, 19.5°E)		0.556	0.067		Dlugosz et al. (2010)
Xiamen, China	2004–2005		(24.4°N, 118.1°E)			0.03–0.21 (0.11)		Yi et al. (2007)
Michigan, USA	1999–2001	30	(42.4°N, 83.0°W)	190	0.30–4.22	BDL-0.118 (0.072)	0.0–0.21 (0.075)	McNeary and Baskaran (2007)
Malaga, Spain	1996–2001		(36.7°N, 4.5°W)		0.51	0.045–0.070	0.11–0.13	Duenas et al. (2004)
South Pole	November 2000–January 2001		(90°S, 102°E)	2841	0.2	0.022 ± 0.0011	0.13 & 0.18	Arimoto et al. (2004)
Egypt	January–December 2002		(28.1°N, 30.6°E)		1.2 ± 0.15			Ahmed et al. (2004)
Alaska	January–March 1996	10	(65.1°N, 147.5°W)		0.22–1.02	0.0–0.152	0.0–0.177	Baskaran and Shaw (2001)
Argonne, Illinois	June 1996–August 1996		(41.7°N, 88°W)					Marley et al. (2000)
Jungfraujoch	January–December 1991		(46.5°N, 8°E)	3450	0.005–0.5			Gaggeler et al. (1995)
Boulder, Colorado	1967–1972		(40°N, 103.3°W)		0.15–0.78	0.002–0.052		Poet et al. (1972)
Milford, Haven			(51.7°N, 5°W)					Peirson et al. (1966)

$^{\$}$above sea level.
BDL = below detection limit.

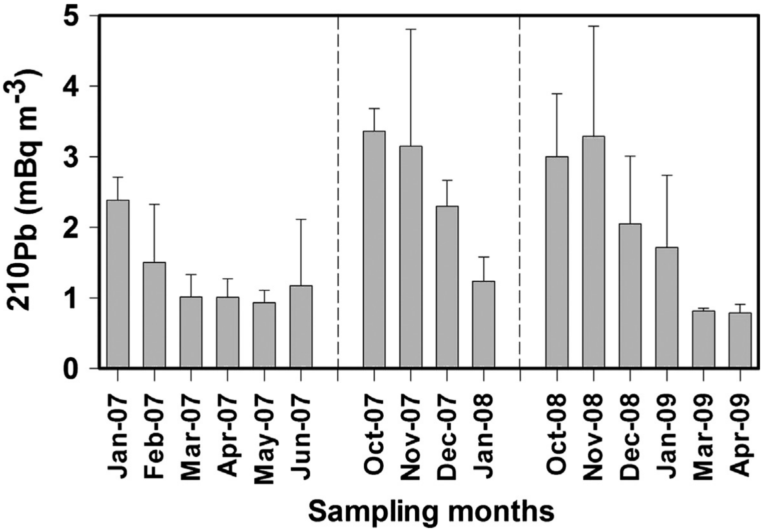

Fig. 3. Average ^{210}Pb activity in urban aerosols during the sampling period. The vertical dash-lines indicate breaks in the aerosol sampling.

seasonal variability, with higher values during the wintertime compared to summertime (the average winter/summer ratio being 1.46) (Singh et al., 2005). It may, thus, be inferred that shallow boundary layer height during the wintertime could lead to an efficient trapping of chemical constituents (Ram et al., 2010), ^{222}Rn and its decay products. In contrast, the time period of March–June is characterised by relatively high ambient temperature and wind speed, which can lead to unstable atmospheric conditions and more convective mixing. These meteorological conditions have led to decrease in concentrations of carbonaceous species and ^{210}Pb activity (Fig. 2). Furthermore, inorganic species such as sulphate, nitrate and ammonium also exhibit a similar seasonal pattern, with lower concentrations in the summer (Ram et al., 2010).

The seven-day back-trajectory analyses are performed for three different types of air masses (Section 2.4) arriving at Kanpur (Fig. 1), and average ^{210}Pb activity associated with these air masses are presented in Table 2. The highest ^{210}Pb activity is observed for the localised air masses (Av: $2.5 \pm 1.2\,\mathrm{mBq\,m^{-3}}$) which is two to three times higher than that associated with air masses from Desert areas and maritime air masses (Table 2). The majority of air masses are of continental origin with relatively low wind speed (Av: $0.4 \pm 0.3\,\mathrm{m\,s^{-1}}$) and are characterised by high ^{210}Pb activity. In contrast, wind patterns in Fig. 1a show the dominance of south-westerly winds originating from the Desert regions of the Afghanistan and the Middle East. These source regions have depleted ^{210}Pb activity. For example, Sayed et al. (2002) had reported that ^{210}Pb concentrations varied from 0.002 to $2.53\,\mathrm{mBq\,m^{-3}}$, with an average value of $0.896\,\mathrm{mBq\,m^{-3}}$ in the atmospheric aerosols collected at Kuwait. More recently, Ali et al. (2011) have reported that ^{210}Pb activities are lower in ambient aerosols (Av: $0.28 \pm 0.15\,\mathrm{mBq\,m^{-3}}$) at Islamabad.

The annual average ^{210}Pb activity in ambient aerosols at Ahmedabad (an urban site) and Mount Abu (a high-altitude site), located in western India, was reported to be $0.9 \pm 0.4\,\mathrm{mBq\,m^{-3}}$ and $0.8 \pm 0.4\,\mathrm{mBq\,m^{-3}}$, respectively (Rastogi and Sarin, 2008). The ^{210}Pb activities were also lower in aerosol samples collected during summer months ($0.33 \pm 0.11\,\mathrm{mBq\,m^{-3}}$ at Ahmedabad and $0.4 \pm 0.2\,\mathrm{mBq\,m^{-3}}$ at Mount Abu, respectively) (Rastogi and Sarin, 2008). The back-trajectory analyses during March – June suggest that air mass originates from oceanic regions (Fig. 1b) and is associated with lower ^{210}Pb activity ($0.9 \pm 0.2\,\mathrm{mBq\,m^{-3}}$) (Table 2). Likewise, air masses from the Desert regions of Afghanistan and the Middle East and oceanic regions are depleted in ^{210}Pb.

3.2. *^{210}Po in aerosols*

The ^{210}Po activity at Kanpur varied from 0.002 ± 0.02 to 0.28 ± 0.04 mBq $\mathrm{m^{-3}}$ (Fig. 6a) and is similar to that reported over Chinese regions, Poland and South Pole (Arimoto et al., 2004; Dlugosz et al., 2010). Dlugosz et al. (2010) had reported that ^{210}Po activity varied from 0.01 to $0.431\,\mathrm{mBq\,m^{-3}}$ in the urban air of the Polish city of Lodz, Poland, during the spring and winter seasons of 2008–2009. However, lower ^{210}Po activity has been also reported in atmospheric aerosols (Poet et al., 1972;

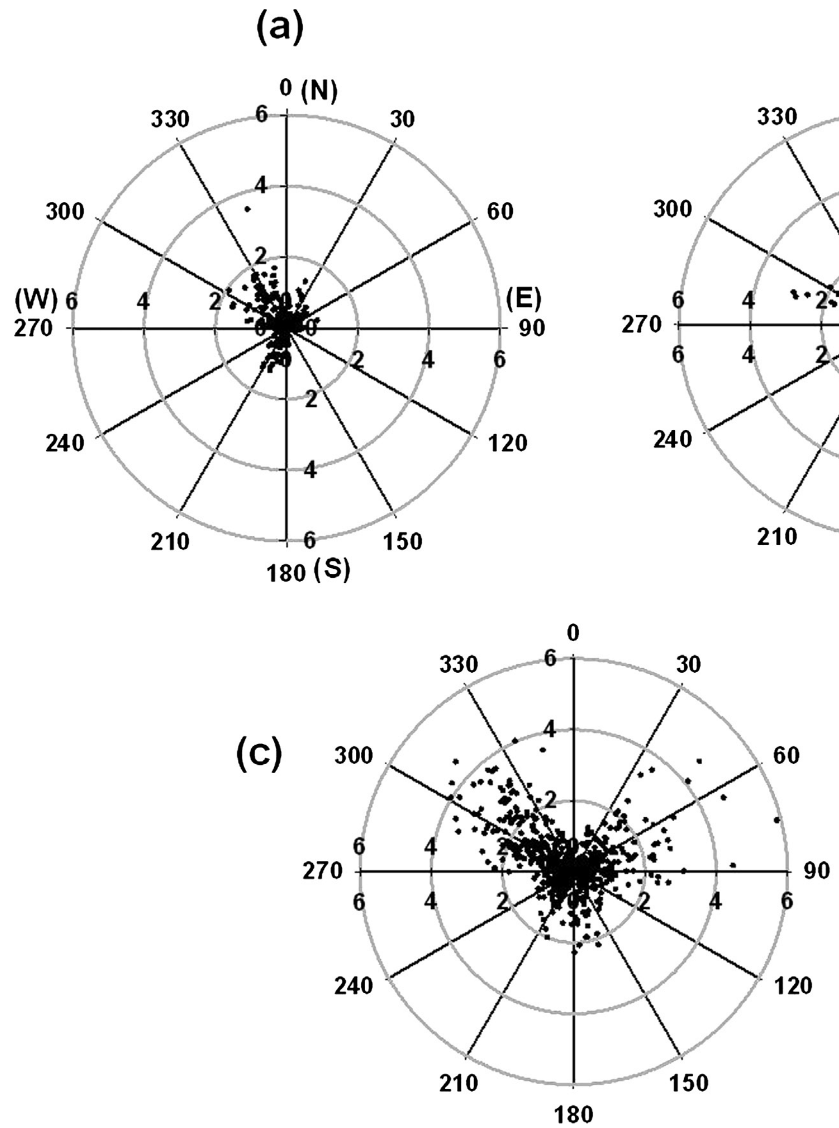

Fig. 4. Seasonal wind-rose plot for the sampling period from January 2007 to April 2009: (a) October–November, (b) December–February and (c) March–June. The acronyms N, E, S and W refer to North, East, South and West directions, respectively.

Duenas et al., 2004; Yi et al., 2007). In a study conducted over southeastern Michigan, ^{210}Po activity ranged from below detection limit to 0.118 mBq m^{-3} in aerosol samples (McNeary and Baskaran, 2007). An inter-comparison of ^{210}Po activity and ^{210}Po/^{210}Pb activity ratio in aerosol samples from different locations is presented in Table 1. The relatively high ^{210}Po activity at Kanpur (Fig. 6) suggests its contribution from other sources (see Section 3.3).

3.3. *Source (s) of ^{210}Po activity at study site*

The temporal variability in ^{210}Po and ^{210}Po/^{210}Pb activity ratio is shown in Fig. 6. The relatively high ^{210}Po activity is a characteristic feature seen during October–November and December–February when an increase in the source strength of biomass burning emissions with a simultaneous increase in abundances of carbonaceous species (EC, OC) and K^{+} is clearly evident (Ram et al., 2010). A maximum value of 0.23 for ^{210}Po/^{210}Pb activity ratio is recorded in the samples collected during the month of April (Fig. 6c).

It is relevant to discuss other sources and direct emission of ^{210}Po to the ambient atmosphere. It is well recognised that volcanic eruption can contribute significantly to atmospheric ^{210}Po, as erupted lavas are characterised by high ^{210}Po/^{210}Pb activity ratio (Lambert et al., 1985). A small mass fraction of volcanic aerosols could, thus, lead to high levels of ^{210}Po in the atmosphere. Likewise, emission

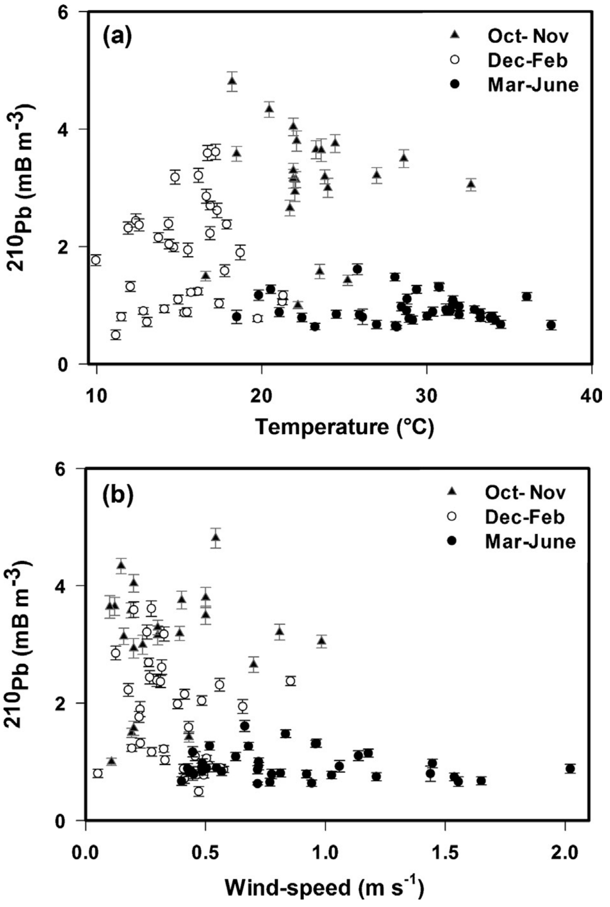

Fig. 5. The relationship between ^{210}Pb activities with (a) ambient temperature and (b) wind speed in aerosols at Kanpur during the sampling period. The data for October–November (post-monsoon), winter (December–February) and summer (March–June) are shown separately. The lower temperature and wind-speed are typical of the wintertime conditions due to lower boundary layer height.

of volatile species containing ^{210}Po from sea-surface and coastal waters (associated with phytoplankton) is suggested as a possible source of ^{210}Po to the atmosphere (Kim et al., 2000). In this study, the sampling site is far away from an active zone of volcanic activity (the nearest one is in Andaman Island); we, thus, rule out any contribution of ^{210}Po from volcanic emissions.

Biomass burning emissions have been suggested as a potential source of ^{210}Po in the atmosphere (Francis et al., 1968; LeCloarec et al., 1995; Nho et al., 1996). Both ^{210}Po and ^{210}Pb enter into plant material through the root uptake of their activity levels present in the soils. The accumulation of ^{210}Pb and ^{210}Po can lead to significant activity levels in plant tissues. Recently, Manigandan (2009) has studied the transfer of natural radionuclides from soil to plants in the tropical forest of Western Ghats in India and reported that the activity levels of ^{210}Po and ^{238}U in plants and soil reflected the impact of the igneous nature of rocks in the study area. Interestingly, this study has reported that the ^{210}Po activity levels are relatively higher than that of ^{238}U in most of the samples (soil as well as plants). Furthermore, the concentration ratio (defined as the activity ratio in plant to that in soil) for ^{210}Po was relatively higher than that for ^{238}U, suggesting greater transfer of Po than U in plants. It was also concluded that the amount of radionuclides in different plant species (by uptake) depends on the substrate concentration, nature of the plants and ageing of the plant. Being volatile in nature, a significant amount of ^{210}Po from plant tissues is released back to the atmosphere during biomass burning emissions. It is well documented that the strength of biomass burning emissions contributes significantly to the aerosol composition during the wintertime in the IGP (Rengarajan et al., 2007; Gustafsson et al., 2009; Ram et al., 2010).

Emissions from coal combustion have been also identified as a dominant anthropogenic source of ^{210}Po in Seoul, Korea (Kim et al., 2005). However, the contribution from coal emissions to carbonaceous aerosols and ^{210}Po is not relevant in the absence of any major coal-based power plants and industrial units located upwind of the sampling site (Kanpur). It is important to reiterate that biomass burning emissions overwhelm the chemical composition of carbonaceous aerosols in the IGP (Rengarajan et al., 2007; Ram et al., 2010). As argued in the preceding sections that ^{210}Pb and ^{210}Po exhibit pronounced seasonal pattern similar to that documented for carbonaceous species (Fig. 2), it is, thus, inferred that biomass burning emissions contribute significantly to atmospheric ^{210}Po during wintertime at the study site.

Table 2. The average ^{210}Pb activity in different types of air masses arriving at Kanpur during the sampling period

Air-mass type	No. of trajectories	Occurrence (%)	Wind speed (ms$^{-1)}$	^{210}Pb (mBq m^{-3})
Desert areas	20	40	0.7 ± 0.6	1.3 ± 0.5
Maritime	11	22	0.9 ± 0.4	0.9 ± 0.2
Localised winds	19	38	0.4 ± 0.3	2.5 ± 1.2

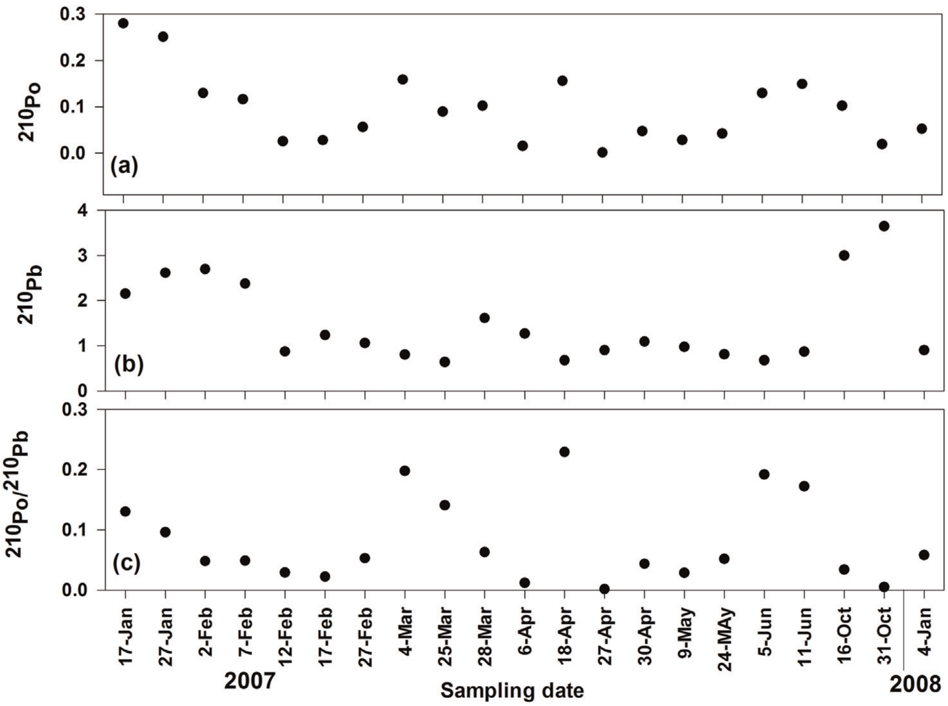

Fig. 6. Temporal plot of activities (in mBq m^{-3}) of environmental nuclides: (a) ^{210}Po, (b) ^{210}Pb and (c) ^{210}Po/^{210}Pb activity ratio at the sampling site (Kanpur) in the Indo-Gangetic Plain.

4. Conclusion and implication

The atmospheric ^{210}Pb and ^{210}Po activities studied in urban aerosols from the IGP suggest large temporal variability, with significantly high activity levels in the wintertime. The characteristic seasonal pattern of biomass burning emissions (dominated by agricultural-waste burning) in the wintertime, a shallow boundary layer height and efficient trapping of pollutants (EC, OC and inorganic constituents) are the dominant factors for significantly high ^{210}Pb and ^{210}Po activities in the IGP.

This study provides an important data-set on ^{210}Pb and ^{210}Po activities for the Global Atmospheric Watch Programme. The ^{210}Pb activity at Kanpur (26.5°N), representing central region of the IGP, is three times higher than the model derived value at this latitude; suggesting the importance of regional lithology (soil characteristics) and environmental factors (meteorology and emission sources) in the model calculations.

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