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Temporal variability in residence time of ambient aerosols using environmental ^{210}Pb

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The regional air quality, atmospheric chemistry and climate change are largely influenced by the chemical composition of ambient aerosols and more importantly, by their residence time on a spatial and temporal scale. The environmental radionuclide ^{210}Pb ($t_{1/2}$ = 22.3 years), injected into the atmosphere by *in situ* decay of its parent nuclide ^{222}Rn ($t_{1/2}$ = 3.8 days) from ground sources, is highly particle reactive and thus serves as an ideal tracer to assess the residence time of anthropogenic and natural aerosols from ground-based sources. We report on the temporal variability in residence time of ambient aerosols studied from an urban site (Ahmedabad) and a high altitude site (Mt Abu) in western India. The residence time of aerosols, ranging from ~2 to 8 days, is predominantly controlled by regional meteorology and high dust abundance (shorter removal time) in semi-arid regions. These observations raise the issue of uncertainty in tracing the source region of atmospheric pollutants based on air-mass back trajectory analyses without knowing their actual residence time for a given time-period over a study region.

Keywords: Ambient aerosols, radionuclides, residence time, temporal variability.

AMBIENT aerosols, either emitted directly in particle form (primary particles), or formed in the atmosphere by physico-chemical processes (secondary particles), originate from a variety of natural (sea salt, dust, volcano, etc.) and anthropogenic (fossil fuel and biomass burning) sources. Although the residence time of ambient aerosols in the lower troposphere is reported to be only about a week, their effects from growing anthropogenic sources are increasingly recognized in the deterioration of air quality, atmospheric chemistry and Earth's radiation budget. The residence time of atmospheric aerosols depends upon various removal processes, e.g. dry deposition (by impaction and sedimentation) and wet deposition (by rain, snow). Detailed information of both spatial and temporal variability in the residence time of ambient aerosols is thus essential to understand their atmospheric transport and removal processes.

Atmospheric ^{210}Pb ($t_{1/2}$ = 22.3 years) together with ^{222}Rn ($t_{1/2}$ = 3.8 days), ^{210}Po ($t_{1/2}$ = 138 days) or ^{210}Bi ($t_{1/2}$ = 5 days) have been used to estimate the residence time of atmospheric aerosols[1–5]. Figure 1 depicts a sche-

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matic diagram for the production and removal mechanism of ^{210}Pb in the atmosphere. ^{210}Pb is a naturally occurring radiogenic nuclide. The source of atmospheric ^{210}Pb is the radioactive decay of ^{222}Rn through short-lived daughters nuclides (Figure 1). ^{222}Rn is continuously emitted from surface soils/rocks to the atmosphere from the decay of ^{226}Ra (decay daughter of ^{238}U decay series, Figure 1). Anthropogenic sources such as coal combustion and nuclear explosion, contribute less than 1% of ^{222}Rn and ^{210}Pb in the atmosphere[6]. ^{210}Pb is formed via gas-to-particle conversion similar to many secondary aerosols (e.g. sulphate, nitrate, organics). Soon after production, ^{210}Pb gets attached to ambient particles and then its fate is similar to aerosols, implying that it is transported in the atmosphere on scale length ranging from local to synoptic and global, and ultimately removed from the atmosphere via dry and wet deposition. Since the half-life of ^{210}Pb is very long in comparison to the typical residence time of aerosols in the lower troposphere, its removal via radioactive decay is insignificant. As most of ^{222}Rn (> 99%) emanates from the Earth's surface, ^{210}Pb is widely used as a tracer of continental air mass over remote oceans[2,7–9].

In this communication, we report ^{210}Pb-based residence time of ambient aerosols, its temporal variability over an urban site (Ahmedabad) and a high altitude site (Mt Abu) located in western India, and its implications in source identification of atmospheric pollutants emitted from ground-based anthropogenic sources.

Details of sampling sites, sample collection and analyses, and the values of ^{210}Pb in each collected samples have been published elsewhere[9]. Briefly, the study sites were located at Ahmedabad (23.0°N, 72.6°E, 49 m amsl) and at Guru Shikar, Mt Abu (24.6°N, 72.7°E, 1680 m amsl). These sites are characterized by similar meteorological conditions and located in the semi-arid region of western India, but are different with respect to altitude and ambience (urban versus remote). The sites are influenced by two types of air mass (polluted continental and marine) derived predominantly by NE and SW winds during November–February and May–August respectively[9,10]. During March–April and September–October, mixed air masses are received due to change in major wind patterns (NE/SE). The average rainfall over the sites is ~ 650 mm, most of which occurs during the SW monsoon. The remaining months experience dry conditions (low humidity, high dust).

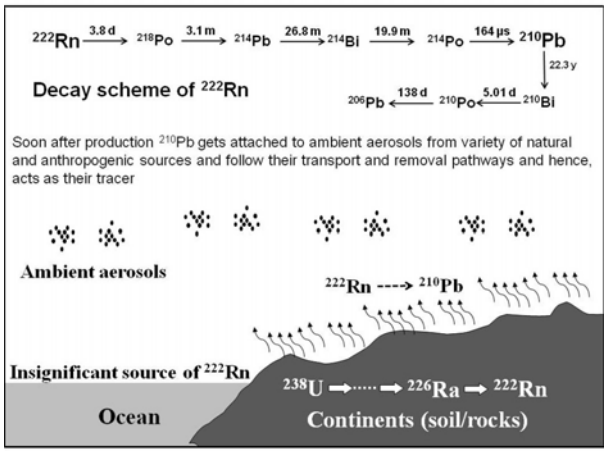

Figure 1. Schematic diagram for production and removal of ^{210}Pb in the atmosphere.

Bulk aerosol or total suspended particulates (TSP) samples were collected using high-volume air sampler during May 2000 to January 2003 over Ahmedabad (n = 89), and during October 2001 to January 2003 over Mt Abu (n = 25). ^{210}Pb in all these aerosol samples was assayed by gamma counting (γ-peak at 46.5 keV measured by HPGe well detector, Canberra Model GCW 2523). The propagated errors (those arising from detector calibration and sample count rates) were less than 10%. The abundances of ^{210}Pb in few aerosol samples, collected during SW monsoon were close to detection limit (< 0.10 mBq m^{-3}).

The concept of estimating residence time based on pairs of radionuclide (^{222}R–^{210}Pb and ^{210}Pb–^{210}Bi) is reasonably straightforward. Under steady-state conditions, production rate of the radionuclide of interest (^{210}Pb in this case) in the atmosphere is balanced by its removal by radioactive decay and dry and wet depositions, i.e.

$$dN_D/dt = \lambda_P N_P - \lambda_D N_D - \lambda_R N_D = 0. \quad (1)$$

Here, $\lambda_P N_P$ is the activity of parent nuclide which is same as the rate of production of daughter nuclide; the $\lambda_D N_D$ is the rate of decay of daughter nuclide, and $\lambda_R N_D$ is the rate of removal ($\lambda_R N_D$) of daughter nuclide from the atmosphere. Here, λ_R (= $1/\tau$) is the first-order rate constant for the removal of aerosols (where daughter nuclide is attached) from the atmosphere by all processes and τ is the residence time. After rearranging eq. (1), the residence time (τ) can be estimated as

$$\tau = 1/\lambda_D \times (A_D/A_P)/[1 - (A_D/A_P)], \quad (2)$$

where A_D (= $\lambda_D N_D$) and A_P (= $\lambda_P N_P$) are the activities of daughter and parent respectively, for the selected pairs of radionuclide. Recently, aerosol residence time has been estimated using solely ^{210}Pb concentration in ambient aerosols, presuming that the decay term ($\lambda_D N_D$) for ^{210}Pb would be negligible as residence time of aerosols is in the order of a few days in comparison to half-life of ^{210}Pb (ref. 9). Therefore, eq. (2) will become

$$\lambda = 1/\lambda_D \times (A_D/A_P). \quad (3)$$

Using eq. (3), we have estimated the residence time of ambient aerosols based on long-term continuous aerosol ^{210}Pb data over Ahmedabad and Mt Abu. In eq. (3), the production rate of ^{210}Pb, i.e. activity of ^{222}Rn (A_P) in the atmosphere (= 1.2 atom cm^{-2} s^{-1}) is well constrained[2], and

A_D is the measured activity of ^{210}Pb atoms in the air column of selected height (for which residence time has to be calculated). The basic assumption in this approach is that the activity of ^{210}Pb is uniform within the air column for a selected height and location. Turekian *et al.*[2] have shown that ^{210}Pb activity decreases with increasing altitude, but remains more or less uniform until ~6 km amsl. Therefore, in the present study, we have selected the lower tropospheric air column up to 6 km for estimating the residence time of lower tropospheric aerosols.

Figure 2 presents the temporal variability in monthly average ^{210}Pb concentrations at the two study sites. ^{210}Pb concentration in aerosols varied from 0.10 ± 0.01 to 1.9 ± 0.20 mBq m^{-3} (avg: 0.73, sd: 0.44) over Ahmedabad, and from 0.25 ± 0.04 to 1.8 ± 0.20 mBq m^{-3} (avg: 0.68, sd: 0.38) over Mt Abu with significant seasonality over both the study sites. However, inter-annual variability was not prominent (Figure 2). The observed concentration values of ^{210}Pb are similar to those reported over other sites in India[11].

The temporal variability in ^{210}Pb could arise due to different environmental factors. During the period of the SW-monsoon (May–August), ^{210}Pb activity was lower than the overall average value. This is attributed to wind regimes over the study region prevailed by strong SW winds that also carry marine air mass from the Arabian Sea (expectedly poor in ^{210}Pb), and efficient wet removal of ^{210}Pb by rain events during this season. Higher ^{210}Pb activity was observed from November to January, a period dominated by NE winds, and ascribed to continental air mass brought by NE winds with near absence of wet removal during this period over the study region. Intermediate concentrations of ^{210}Pb were observed during February–April and September–October when transition between major wind regimes (NE and SW) occurs. The period from February to March is also spring time with maximum vertical mixing of air masses[9]. Thus, lower tropospheric air mass (enriched in ^{210}Pb) gets diluted with upper tropospheric/stratospheric air mass (poor in ^{210}Pb) resulting in lower ^{210}Pb concentration in the lower troposphere. The period from April to June is characterized by high ambient temperature and therefore, convective vertical mixing of air masses remains significant, leading to lower ^{210}Pb concentrations. Also, the mineral dust abundance is high (~ 60% of TSP) throughout the year over the study region with further higher concentration (~ 70%) during April–June due to dry conditions[12]. The mineral dust particles, which have shorter lifetime due to coarser size, are the main scavengers of ^{210}Pb over the study region and thus, reduce ^{210}Pb concentration in the atmosphere. Such an observation suggests that the type of air mass (continental vis-á-vis marine, lower troposheric vis-á-vis upper tropospheric) and size of aerosols (coarse/fine) strongly affect ^{210}Pb abundance, in addition to wet removal over the study region.

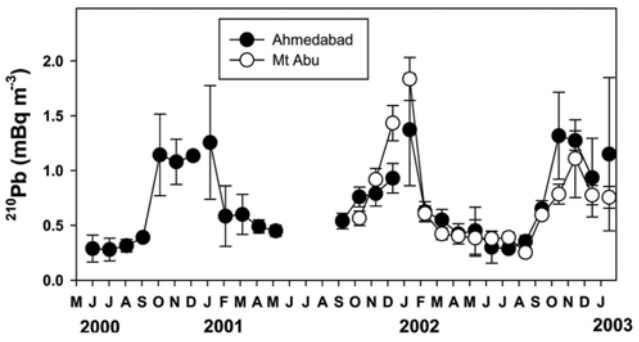

Figure 2. Monthly average ^{210}Pb concentration in aerosols over Ahmedabad and Mt Abu during the study period. Error bars represent standard deviation on the monthly average.

As discussed earlier, pairs of radionuclide from ^{222}Rn daughters are often used to estimate the residence time of atmospheric aerosols. Using ^{210}Po–^{210}Pb pair, McNeary and Baskaran[5] have reported the residence time of aerosols varying between 0 and 61 days, with a mean value of 22 days over Michigan, USA, whereas Duenas *et al.*[6] have calculated the mean residence time of 31 days at Malaga, Spain. Kim *et al.*[13] compared the residence time estimated from ^{210}Po–^{222}Rn and ^{210}Po–^{210}Pb pairs and attributed the longer residence time obtained from the latter pair to the excess ^{210}Po derived from surface waters either by sea spray of the microlayer or by gas exchange. According to Moore *et al.*[14], the mean residence time of aerosols in the troposphere is less than 7 days and increases with altitude within the troposphere. The residence time of aerosols cannot be a fixed number because atmospheric aerosol abundance depends on their primary emission sources, secondary formation and regional meteorology.

Figure 3 depicts the estimated residence time of ambient aerosols over Ahmedabad and Mt Abu based on all the monthly average ^{210}Pb concentrations (error bars indicate the standard deviation on the mean value of residence time for a given month for different years). The residence time of aerosols varied between ~ 2 and 8 days over both the sites, with higher value during winter and lower during SW monsoon (Figure 3). Relatively low residence time during the SW monsoon (~ 2 days) is attributed to efficient wet scavenging of atmospheric constituents via frequent rain events. During winter, the residence times of lower tropospheric aerosols at both the sites were between 6 to 8 days, similar to those reported in the literature[1,3,4,15]. Higher residence time over the study sites during winter was attributed to meteorological conditions such as lack of rain, weaker winds and stagnant atmosphere with respect to vertical mixing[9]. Residence times of aerosols over both the sites are similar because both sites are located in the same region and characterized by similar lithology and soil type, and obviously have similar meteorological conditions. Under steady-state conditions, eq. (3) also suggests that the enhanced concentration of ^{210}Pb in aerosols directly reflects their ageing.

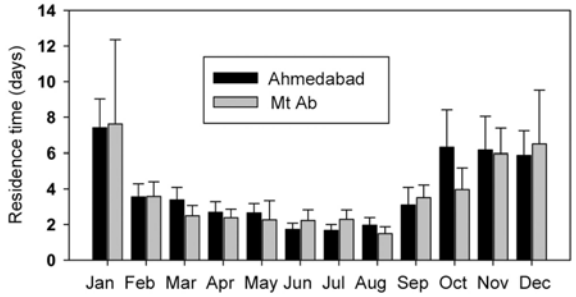

Figure 3. Monthly average residence time of ambient aerosols over Ahmedabad and Mt Abu based on ^{210}Pb abundance in aerosols. Error bars represent the standard deviation on monthly averaged residence time.

Though this method can be used to estimate residence time of ambient aerosols, it has few limitations. The flux of ^{222}Rn shows significant spatial and temporal variability depending upon ^{226}Ra content of topsoil, soil porosity and meteorology of a particular region[16,17]. In our approach, assumption of constant ^{222}Rn emission flux could introduce some error, but this is minimized as it is used with monthly averaged ^{210}Pb because corresponding monthly average ^{222}Rn flux is expectedly much less variable. Further, sample-to-sample non-uniformity in size distribution of aerosol particles can affect ^{210}Pb concentration as fine particles can scavenge radiogenic ^{210}Pb more efficiently than coarse particles due to higher surface/volume ratio of finer particles. Also, the estimated residence time over the sites dominated by marine air masses can be considered as a lower limit because ^{222}Rn emission from the ocean is negligible (1%).

The residence time of ambient aerosols over an urban site (Ahmedabad) and high-altitude station (Mt Abu) varied between ~ 2 and 8 days, with higher values (6–8 days) during winter and lower (~ 2 days) during SW monsoon period. Our observations suggest that regional meteorology (especially precipitation, wind direction, mixing of air masses), and higher abundance of mineral dust control the aerosol residence time over these two study regions. It is suggested that the use of aerosol residence time is more appropriate while identifying the source region of atmospheric pollutants based on air-mass back trajectory analyses. It is also suggested that the enhanced concentration of ^{210}Pb in aerosols directly reflects their ageing.

Seven days air-mass back trajectory analysis is often performed while identifying the source region of atmospheric pollutants derived from various anthropogenic sources, which is implicitly related to the designing of air pollution mitigation strategies. This study suggests that 7 days back trajectory analysis may not always be required. The number of days for which air-mass trajectory analysis is valid shall be chosen based on the residence time of ambient aerosols in a given month, as residence time shows significant variability (e.g. ~ 2–8 days in this study). This approach, which uses ground-based tracer (^{210}Pb), shall reduce the uncertainty associated with identification of source region of air pollutants emitted from continental anthropogenic sources. There are many sites in the world where long-term data on ^{210}Pb are available through Global Atmospheric Watch programme and our simple method of estimating the aerosol residence time based on ^{210}Pb activity should serve as a representative approach.

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