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journal or publication title	Proceedings, International Symposium of the Kanazawa University 22st-Century COE Program
volume	1
page range	161-166
year	2003-03-16
URL	<a href="http://hdl.handle.net/2297/6387">http://hdl.handle.net/2297/6387</a>

## Fate of Polycyclic Aromatic Hydrocarbons and Radionuclides through Loess over Pan-Japan Sea Area -Reaction, Transportation and Deposition-

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**Abstract** - Loess could play a critical role in many processes either indirectly (e.g., climate) or directly (e.g., health). Due to high reactivity of loess, the reaction and transport of pollutants such as polycyclic aromatic hydrocarbon (PAH) and radionuclides might be associated with constituents of loess in troposphere over Pan-Japan Sea area. For kick-off of our research to understand the fate of the pollutants, characteristics and chemical reactivity of loess were reviewed, and research plans of loess were proposed.

### I. Introduction

The atmosphere of the Earth is a colloidal system that contains liquid and solid aerosol particles besides gas-phase components. Aerosol particles, i.e. loess, are ubiquitous and play an important role in the physics and chemistry of the atmosphere, especially in the troposphere. For example, high concentrations of particles can cause serious visibility degradation; some particle types are notable for their contribution to atmospheric acidity, whereas other types are important because of their health effects. On the other front, aeolian dust fluxes and their temporal variations have been studied intensively during the last two decades because of the possible importance of dust to global nutrient and marine biogeochemical cycles [1] and its utility as a paleoclimatic indicator of the aridity of continental interiors [2], especially as a proxy record of variations in Asian monsoon climate [3-6].

Loess falldown from the inland arid in central Asia is called "Kosa (=yellow sand)" in Japan. Frequent dust storms in the arid area of central Asia result in increased delivery of loess to the north Pacific via the prevailing westerlies [7], as well as to the Loess Plateau in China [8]. Dust outbreaks occur in the Asian deserts during spring resulting in seasonally elevated aerosol concentrations as for downwind

as Hawaii and the north-western United States [9]. About 80% of the annual aerosol mass deposition on Mauna Loa is due to mineral dust from Asian sources [10].

It might be true that the flight of loess itself plays an important role in Pan-Japan Sea area as mentioned above. However, from the viewpoint of the material cycle, the role of loess as a media of reaction and material transportation in troposphere is more important than the flight of it due to their high chemical affinity. It was well documented that mineral aerosol particles are mixed with aerosols from pollution sources in Eastern Asia [11], and the originally alkaline aerosol is neutralized by acidic sulfates [12].

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous and have been identified in a variety of soils, natural waters and wastewaters. Although they also occur naturally, high levels of PAHs are mainly attributed to anthropogenic origin. They are also emitted into the atmosphere from numerous combustion process, e.g., traffic, domestic heating, biomass burning, etc. Potential toxic effect of PAHs, that has gained attention in recent years, is endocrine disruption. PAHs and their metabolites have been shown to influence endocrine functioning, although the evidence is limited because of only few PAHs have been studied so far.

Some of radionuclides (naturally occurring, i.e.,  $^7\text{Be}$ ,  $^{10}\text{Be}$ ,  $^{210}\text{Pb}$ ,  $^{222}\text{Rn}$  and anthropogenic, i.e.,  $^{85}\text{Kr}$  and  $^{90}\text{Sr}$ ) have served as ideal tracers because of their simple behavior, nonreactive properties, and the availability of a vast number of radionuclide measurements worldwide. Scientists have used and analyzed these measurements to improve their model calculations for further understanding atmospheric circulation, deposition or removal processes, and the transport of environmental pollutants. The global distributions of the source-sink terms of these radionuclides by latitude, longitude and altitude are well known and extensively useful indicators of their transport.

Goal of our research is to show which particle types are globally or regionally important, their makeup, how they interact with PAH and radionuclide in the atmosphere, how they transport over Asian continent and Pan-Japan Sea area and deposit on Japan Island, and their principal effects on the environment in Pan Japan sea area. For kick-off of our research to understand the fate of the pollutants, characteristics and chemical reactivity of loess were reviewed, and research plans of loess were proposed.

## II. Loess characteristics

### A. Main source regions

Loess formation over continents is caused by the action of wind on the soil surface. The occurrence of dust storms in a given region varies seasonally; in general, the monthly occurrence of dust events is higher during the spring and summer time than in fall and winter [13]. Aeolian dust is generated primarily in arid or semiarid regions. The most active sources of loess plumes are distributed within a band that includes North Africa, the Middle East, and Central Asia. Mineral dust has been considered a natural, "background" component of tropospheric aerosols. However, human activity changes the flux of soil particles into the atmosphere. Changing land use such as deforestation and over-cultivation, particularly in the Sahel region of Africa, were estimated to result in a twofold increase in the global flux of the atmospheric mineral dust burden [14]. However, this view has been challenged [15], who found it difficult to make a deconvolution of the effects in changing land use and natural, interannual climate variability in African dust transport.

### B. Composition

Owing to their different formation mechanisms, the chemical composition of fine and coarse particles, and continental and marine aerosols are substantially different (Fig. 1). Depending on their origins, particles also differ in shape and physical state. In Pan-Japan Sea area, the loess comes from continental region first and through Japan Sea. In this case, the composition of loess should be changed during the flight from continent via Japan Sea to Japan Island. It is not difficult to imagine that the complex reactions have probably occurred, because the chemical condition is drastically changed from continent to marine regions.

### C. Mineralogy

Although the amount of continental dust (usually inferred from the Al content of the aerosol) is continuously monitored at several locations [13], the mineralogy is not. Therefore, we do not know much about the seasonal variability of the dust mineralogy. XRD data are available from several regions; however, these studies can be regarded as one-time experiments that provide snapshots of the

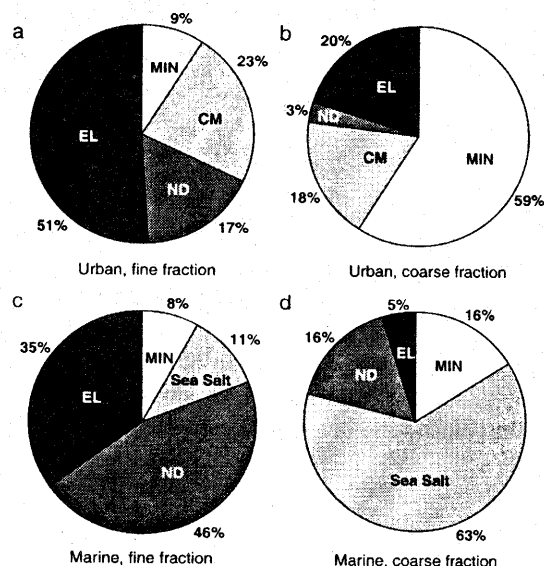


Fig. 1. Selected examples of fine vs. coarse and urban vs. marine aerosol compositions [23]. (a) and (b) are representative urban aerosol samples from Vienna; (c) and (d) are remote marine aerosol samples from Chili, Ogasawara Islands. MIN: mineral; CM: carbonaceous matter, ND: not determined; EL: electrolytes (including  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{Na}^+$ , and  $\text{K}^+$  in (a) and (b), and  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and non-sea-salt  $\text{Na}^+$  in (c) and (d)). The size cut between fine and coarse particles is at 2  $\mu\text{m}$  diameter.

mineralogy of plumes originating in major dusty areas of the Northern Hemisphere. Not surprisingly, the major constituents of dust plumes are common rock-forming minerals, including quartz, clay minerals (illite, kaolinite, chlorite, smectite, and so on), mica, feldspars, and calcite. In general, clay minerals are enriched in the longer they stay in the air.

Fig. 2 shows XRD patterns of the loess (yellow) and paleosols (red) from Loess Plateau near Xian, China. The loess and paleosols have been considered to be a source of "Kosa" in Japan. From the XRD patterns, the loess consists of quartz and calcite as major constituent, and illite, chlorite and kolinite as minor constituent. On the contrary, sample from the paleosol mainly consists of quartz with minor clay minerals. Environmental scanning electron microscope (ESEM) images of the paleosol show that the presence of tiny clay particles was observed besides quartz which is containing as relatively big particle and dominant in terms of volume in the loess and paleosol. ESEM is powerful tool for recording the images at the same condition of atmosphere. For example, the image was recorded at 750Pa at 3  $^\circ\text{C}$ . By using ESEM, we can get real images of aerosol particles (shapes, morphology and association) in atmosphere.

Fe-bearing mineral particles have another important constituent for global cycle of the Earth: in large regions of the ocean, Fe is the limiting nutrient for phytoplankton [16],

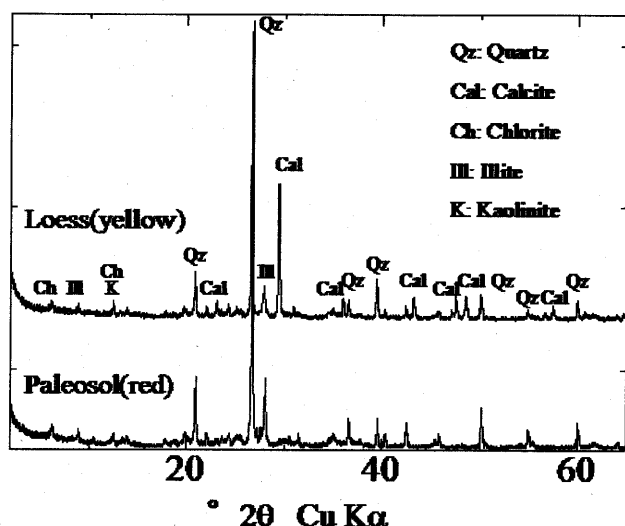


Fig. 2. XRD patterns of loess and paleosol from Loess Plateau near Xian, China.

and aerosol particles supply the Fe that is necessary for the growth of organisms at the base of the food chain [17]. Only scattered information is available about Fe-bearing minerals in the aerosol; for example, Fe was found to reside in smectites and in goethite above the North Atlantic, and also in fly ash particles [18-19]. The bioavailability of Fe is determined by its speciation; systematic studies are needed to determine the speciation of Fe in dust particles. Fe-sulfate interaction is also important for Fe cycle itself but also sulfate cycle. Ferrihydrite and schwertmannite, iron-oxyhydroxide and iron-oxy-hydroxy-sulfate minerals and good scavenger of hazardous cations and oxyanions such as lead, cadmium, chromium, arsenic and selenium, are key phase containing Fe and sulfate in atmosphere and subsurface of the Earth [20-22]. Trace metals (other than Fe) can also influence chemical reactions in aerosol water and cloud drops, and some represent a health hazard. Al, Ti, and Mn are generally considered natural mineral components of the aerosol and mostly occur in the coarse fraction, whereas Pb, Zn, Cu, Cr and Cd are typically of anthropogenic origin, and occur in particles with diameters less than 1  $\mu\text{m}$ .

#### D. Sea salt

In terms of mass, sea salt particles are the most abundant aerosol species in the marine atmosphere; they are also transported to great distances above the continents. Even though the formation mechanism and the basic physical and chemical properties of sea salt aerosol particles have been known for decades, the details of their atmospheric reactions and their climate effects are still receiving much attention. The initial composition of sea-salt aerosol particles is a direct consequence of their production mechanism. They contain dissolved ions in the same relative amounts as seawater.

#### E. Sulfates

Sulfate particles are typically composed of  $(\text{NH}_4)_2\text{SO}_4$  in the continental aerosol because  $\text{NH}_3$  is abundant above the continents. In the marine aerosol the composition of sulfates varies between  $\text{H}_2\text{SO}_4$  and  $(\text{NH}_4)_2\text{SO}_4$ . Sulfate particles can be crystalline or liquid while in the atmosphere, depending on temperature and relative humidity (RH). Pure and dry  $(\text{NH}_4)_2\text{SO}_4$  particles grow nearly linearly up to 81% RH; such particles may be considered solid phases that are coated by water. At 81% RH,  $(\text{NH}_4)_2\text{SO}_4$  particles deliquesce, i.e., they become solution droplets. During dehydration, the same particles remain in a metastable liquid state until they effloresce at about 30% RH. Since most  $(\text{NH}_4)_2\text{SO}_4$  particles in the lower troposphere experience RHs that exceed 81%, they are deliquesced droplets most of the time, owing to the hysteresis loop between hydration and dehydration curves [23].

#### F. Carbonaceous particles

In terms of their concentration and climate effects, the aerosol particles composed mainly of carbon are as important as sulfates above the continents. Organic compounds are emitted into the atmosphere by both natural and anthropogenic sources. Up until recently the organic fraction of the loess was not taken into account, mainly because experimental data were lacking. It is now well known that organic carbon compounds account for 10 to 70% of fine aerosol mass. The sources of fine aerosol organic carbon, called "shoot" are primarily biomass burning, fossil fuel combustion and biological material, and secondarily oxidation of anthropogenic volatile organic carbon (VOC) and oxidation of biogenic VOC. Since soot particles are the strongest absorbers of solar radiation among all atmospheric aerosol types, often their optical properties are emphasized and they are referred to as black carbon particles. Soot particles have typical morphologies and microstructures. Combustion produces hydrocarbons that condense immediately at the source and form 20 to 50 nm large, solid spherules with fingerprint structure.

### III. Chemical reactivity of atmospheric fine particles

The solid phases involved in chemical reaction in atmosphere are typically composite materials, that is, they consist of mixtures of various minerals and organic debris with a wide range of intrinsic chemical properties. Weathering reactions give rise to the reaching of surface layers of minerals and deposition of extremely fine-grained, poorly crystalline minerals. The process also leads to the formation of organic and hydroxypolymer (i.e., amorphous Al or Fe hydrous oxide) coatings on mineral grains. The composite materials of natural system are observed. In many cases, the surface chemical properties of natural materials are dominated by secondary minerals and coatings such as clay minerals, and organic and hydroxypolymer, which usually constitute only a minor fraction of the whole sample.

There are three main reasons why clay minerals are

important solid phase components in natural systems [24]. The first is that they have large specific surface areas (between 10 and 800 m<sup>2</sup>g<sup>-1</sup>) in comparison to other solid phase components. Second, this expansive surface often has an electrical charge associated with it due to isomorphic substitution, defects, and broken edges. This charge results in the accumulation of inorganic and organic cations and is responsible for the high water retention capacity in many types of clay. Finally, naturally occurring clay particles are often coated by amorphous oxides and humic materials and serve as efficient templates for secondary solid phase. As a result, the presence of even small amounts of these fine-grained materials on a mass basis will exert a large influence on the behavior of organic and inorganic solutes in soil and subsurface environments. Thus, in terms of chemical reaction in atmosphere, it is necessary to consider it by the chemical affinity and the reactive surface of constituent particles rather than their particle size.

#### IV. Records of the atmospheric inputs of loess and pollutants

The East Asian monsoon paleoclimate has been well documented by loess-paleosol sequences in the Loess Plateau of China. Several proxy indicators have been proposed to reflect variations of monsoon climate through quasi-independent pathways in the continental and atmospheric systems. For example, grain-size variations of the coarse quartz fraction in loess indicate the relative wind strength of the dust-bearing winter monsoon. Magnetic susceptibility of loess paleosol sequences responds to the intensity of pedogenesis and records the circulation change of the summer monsoon. However, each of these monsoon proxies needs to be verified by other indices that are independent and vary synchronously. Thus, it is necessary to develop new proxy indices. Some geochemical parameters such as the Ge/Si ratio of opaline silica [25] and strontium isotopic ratio of marine carbonates [26], Rb/Sr ratios in the loess-paleosol sequences [27], iron phases [28], have proved to be important in reconstructing the history of continental weathering and long-term Asian monsoon evolution. However, in order to estimate loess flux from the sedimentary record, it is necessary to distinguish subcomponents within the detrital component, specify their origin, and estimate their contents.

Soils and sediments are universal sinks for these pollutants. Under certain sediment conditions, it is possible to determine the historical record of deposition of contaminants in sediments. Until now, the historical records of PAH pollution was largely derived from the analysis of lake and sea sediments [29-30]. If no biodegradation and no transport of PAH after deposition would be recognized, it is available to obtain the data for the reconstruction of the historical input variation into the sediments. The historical input variation is closely related to the history of anthropogenic effects on environment of Pan-Japan Sea area.

#### V. Rationales and strategies for the study of loess as reactors, transporters and recording media

It is important to recognize that aerosol cycles are physical processes linked with one or more atmospheric chemical cycles. In a sense, the physical mixing of aerosol cycles integrates elemental cycles and enables chemical interactions to occur which otherwise might not occur. It is precisely this integration of physical and chemical aspects which suggests that aerosol cycles should be considered as a category separate from those for individual elements. Just as in the case of elemental cycles, aerosols have sources and sinks; aerosol burdens can be estimated, and equations for mass continuity can be written. In addition to the global cycle, loess in lake and marine sediments has good record, although it is very difficult to distinguish the sediments of the aerosol origin and those of continental erosion origin.

Research over the past decade or so has shown that the use of single measures is unsatisfactory because aerosols are so size dependent and because aerosols are so chemically heterogeneous; instead, multiple, concurrent measurements must be made. In this context, the aerosol is a good research object challenging in 21<sup>st</sup> century COE program in which researcher in various fields participates. Here, we will consider reasons and strategies for studying loess in study on environment in Pan-Japan Sea area according to [31]. As mentioned above, many of reasons are based on a need to understand the effects of loess on environment in Pan-Japan Sea area. The understanding and integration of data are urgent issue because environment in Pan-Japan Sea is a problem in a lot of countries where economic circumstances and the policy are considerably different. As a matter of convenience, these arguments will be listed separately.

##### A. *The study of loess for their own physical and chemical properties*

The composition and mineralogy of loess, residence times, major sources and sinks and size distributions are justifiable to seek fundamental information about loess.

##### B. *Understanding the role of loess in geochemical transport in natural cycles*

The actual pathways by which materials are chemically transformed, transported, and added into and ultimately removed from the atmosphere depend to a degree on the existence of the loess. Simply, particles of an inert material are lifted into the atmosphere, transported by winds, and eventually deposited. However, the behavior of loess in nature is not so simple because it is containing very a large number of reactions during the transportation. Especially, in the environment of Pan-Japan Sea area, there is Asian continent, big cities with pollutions, many countries and, of course, Japan Sea. It is more complicated for understanding the behavior of loess than that only over continent. Data are

scarce for most remote regions. Few measurements exist for very large areas of Asia. Over the Japan Sea there are almost no data for vast areas in Japan Sea where is sensitive region for the environment of Pan-Japan Sea area and Japan.

### C. Health and ecological effects of loess

Anthropogenic activities are clearly responsible for a large increase in the variety of hazardous aerosols which man must contend. Aerosol components can be health hazard is dependent on the ability of the aerosol to penetrate the respiratory system. Natural processes can also dangerous, for example, viral and bacterial disease agents, radioactive radon daughter products, and irritant smokes and fumes from vegetation fires and geothermal sites. Thus, the toxicity of various compounds dispersed in the aerosol phase is being actively studied, and interesting results have been found. On the other hand, atmospheric chemist are required for studies of ambient aerosol with the objective of identifying potentially hazardous aerosols and of reaching some understanding of the processes affecting their generation, dispersion, transportation, and removal from the atmosphere.

PAHs are mutagenic air pollutants formed as by-product of combustion. After formation and emission, these compounds partition between the gas phase and atmospheric aerosols. The environmental fate of PAH depends, in part, on their distribution between the gas and particulate phases and among particle size fractions. Particle size affects the removal rate of the associated PAH from the atmosphere by dry and wet deposition. However, a particle size alone is not an effective factor. Surface chemistry of the particles containing in loess is important, because the dominant material composition is quite different between the small and large particles. Authors of the present paper believe the humic acids, clay minerals, and metal oxides with low crystallinity are effective phases for the fate of PAH.

In Pan-Japan Sea area, there are many nuclear reactors for power generation and research. Once the nuclear reactor has a terrible accident, this leads the disaster to the environment of an-Japan Sea area. In this context, the radionuclides are a key index, and the monitoring of radioactivity in the fraction of loess for the prompt correspondence of the environmental changes. Some of radionuclides (naturally occurring, i.e.,  $^7\text{Be}$ ,  $^{10}\text{Be}$ ,  $^{210}\text{Pb}$ ,  $^{222}\text{Rn}$  and anthropogenic, i.e.,  $^{85}\text{Kr}$  and  $^{90}\text{Sr}$ ) have also served as ideal tracers because of their simple behavior, nonreactive properties, and the availability of a vast number of radionuclide measurements worldwide.  $^7\text{Be}$  and  $^{10}\text{Be}$  are produced by cosmic-ray interactions in the upper troposphere and the lower stratosphere.  $^{222}\text{Rn}$  is exhaled from the earth's land surface as a result of uranium decay in soil.  $^{210}\text{Pb}$  is produced in the lower troposphere from the decay of  $^{222}\text{Rn}$  in the atmosphere. Most of the  $^{85}\text{Kr}$  in the atmosphere is released during nuclear fuel reprocessing.  $^{90}\text{Sr}$  is the fallout product of nuclear weapon tests. Nuclides of  $^7\text{Be}$ ,  $^{10}\text{Be}$ ,  $^{210}\text{Pb}$  and  $^{90}\text{Sr}$  attach themselves to submicron-size aerosol particles, and, therefore act as aerosol-borne tracers in the atmosphere.  $^{222}\text{Rn}$  and  $^{85}\text{Kr}$ , chemically and physically

inert, act as noble gases of air tracers.

### D. Strategies for the study of loess

One strategy is to segregate loess with the same origin and pathway into different types. The simplest subdivisions include background, continental (before and after passing through industrial cities), maritime (over Japan Sea), stratospheric aerosols and loess in marine and lake sediments. These loess are identified by their relationship to a source (and thus to a geographic location) and of course, to a cycle. Good samples collected simultaneously from a series of reaction pathway from continent to Japan are necessary to understand the fate of PAH and radionuclides. For this, some of indices to specify the source and the network of the systematic sampling stations are necessary. The ultimate sampling system is to utilize the minimum number of stations and the minimum sampling time that will be sufficient to minimize the variance of the measured quantity, which is itself a function of space and time. Aorian dust storms, "Kosa", are appropriate events to minimize the sampling stations.

To specify the source, compositions of stable isotopes, minerals and contaminants in the individual sample will be measured by direct observations using laser abrasion inductively coupled plasma mass spectroscopy (LA-ICP-MS) and transmission electron microscopy (TEM) with either energy-dispersive X-ray spectrometry (EDS) or electron energy-loss spectroscopy (EELS). Sequential extraction method will also be applied to the samples to determine which phase is the most effective for the sorption of PAHs and radionuclides. In addition, atomic force microscopy (AFM) and ESEM are useful for measuring particle sizes and morphologies under ambient conditions, although the particles are not in the air but on a collection surface. Both microscopies can be put to use in the study of the hygroscopic behavior of individual particles, by measuring size changes in an environmental cell under precisely controlled relative humidity conditions. If it is necessary, we can measure the XRD patterns under precisely controlled relative humidity conditions.

Lake sediments are also useful to get the aeolian dust fluxes and their temporal variations, and a proxy record of variations in Asian monsoon climate and human activity. One of authors (K. K.) already obtained huge amount of data concerning particle sizes of sediments with ages. So it is easy to start the study about loess in the sediments when we use the cores from the sediments. Short-term direct sampling of loess will also be planed to collect loess itself from the repository of Aeolian dust in some lakes.

### Acknowledgements

This research was supported by 21st-century COE (Center of Excellence) program of the Japanese Ministry of Education, Culture, Science and Technology (MEXT).

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