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REPLY

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Key Point:

- Central Asian Deserts are the major dust sources in the Philippine Sea

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Reply to Comment by Xu et al. on "Sr-Nd isotope composition and clay mineral assemblages in eolian dust from the central Philippine Sea over the last 600 kyr: Implications for the transport mechanism of Asian dust" by Seo et al.

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Abstract Against Xu et al. (2016), who argued that East Asian Desert (EAD) dust that traveled on East Asian Winter Monsoon winds dominates over Central Asian Desert (CAD) dust in the Philippine Sea with presentation of additional data, we reconfirm Seo et al.'s (2014) conclusion that CAD dust carried on the Prevailing Westerlies and Trade Winds dominates over EAD dust in overall dust budget of the central Philippine Sea. The relative contribution of dust from EADs and CADs using clay mineral composition should be evaluated with elimination of mineralogical contribution from the volcanic end-member which is enriched in kaolinite and overestimate the contribution of EAD dust.

1. Introduction

We welcome this opportunity for additional discussion on the provenance and transport mechanism of Asian dust in the Philippine Sea. Secular variations in mass flux and composition of Asian dust delivered to the marginal seas and remote Pacific have been of interest to understand response of atmospheric circulation system and surface processes of Asian interior to climatic and tectonic oscillations of varying time scales [*Hovan et al.*, 1991; *Li et al.*, 2011; *Rea and Hovan*, 1995]. Especially, dust flux variations in the East Asian marginal seas have been extensively studied to comprehend the East Asian Winter Monsoon (EAWM) variability on millennial to glacial-interglacial climatic cycles [*Jiang et al.*, 2013; *Ming et al.*, 2014; *Wan et al.*, 2012; *Xu et al.*, 2012]. These studies are based on the premise that the East Asian marginal seas beneath the path of the EAWM winds are dominated by dust from the East Asian deserts (EADs; Ordos, Badain Jaran, and Tengger Deserts) [*Shi and Liu*, 2011] (Figure 1). Contrary to this general conception, *Seo et al.* [2014] reported a mineralogical evidence that dust from Central Asian Deserts (CADs; Taklimakan desert and Qaidam Desert, Figure 1) has prevailed in dust budget over EAD dust in a central Philippine Sea site (PC 631; 12°30′N, 134°60′E, 3728 m water depth; Figure 1) for the last 600 kyr and emphasized important role of prevailing wind system (e.g., Westerlies and Trades) in transport of Asian dust to the Philippine Sea. Against this conclusion, *Xu et al.* [2016] argue that EAD dust that traveled on EAWM winds dominates over CAD dust with presentation of additional data from wide region of the Philippine Sea Basin.

2. Sr-Nd Isotope Composition

As pointed out by *Xu et al.* [2016], a combined use of ε_{Nd} and $^{87}Sr/^{86}Sr$ of inorganic silicate fraction is a powerful tool in defining potential sources of eolian dust at a site of interest [*Chen et al.*, 2007; *Li et al.*, 2011; *Nakai et al.*, 1993]. It has been also widely used to evaluate relative contribution of crustal (e.g., Asian dust) and volcanic components using a theoretical mixing relation between two end-members on a ε_{Nd}-$^{87}Sr/^{86}Sr$ cross plot because of distinctive ε_{Nd} and $^{87}Sr/^{86}Sr$ compositions of these two components [*Jiang et al.*, 2013; *Xu et al.*, 2015] (Figure 2a). However, it is often challenging to identify a specific source region of mineral dust, especially between CAD and EAD sources, among many deserts distributed in Asian interior, because of partial overlapping of dust composition in these sources. For example, dust from CADs and Badain Jaran/Tengger Deserts (hereafter termed "BT," classified to EADs) shares the common data field in a ε_{Nd}-$^{87}Sr/^{86}Sr$ cross plot (Figure 2a). Thus, it is not possible to distinguish CAD and BT dust with ε_{Nd} and $^{87}Sr/^{86}Sr$ compositions. In case of PC631, dust contribution from the Ordos is probable in a ε_{Nd}-$^{87}Sr/^{86}Sr$ cross plot (Figure 2a). However, the relative importance of dust contribution between EADs and CADs cannot be

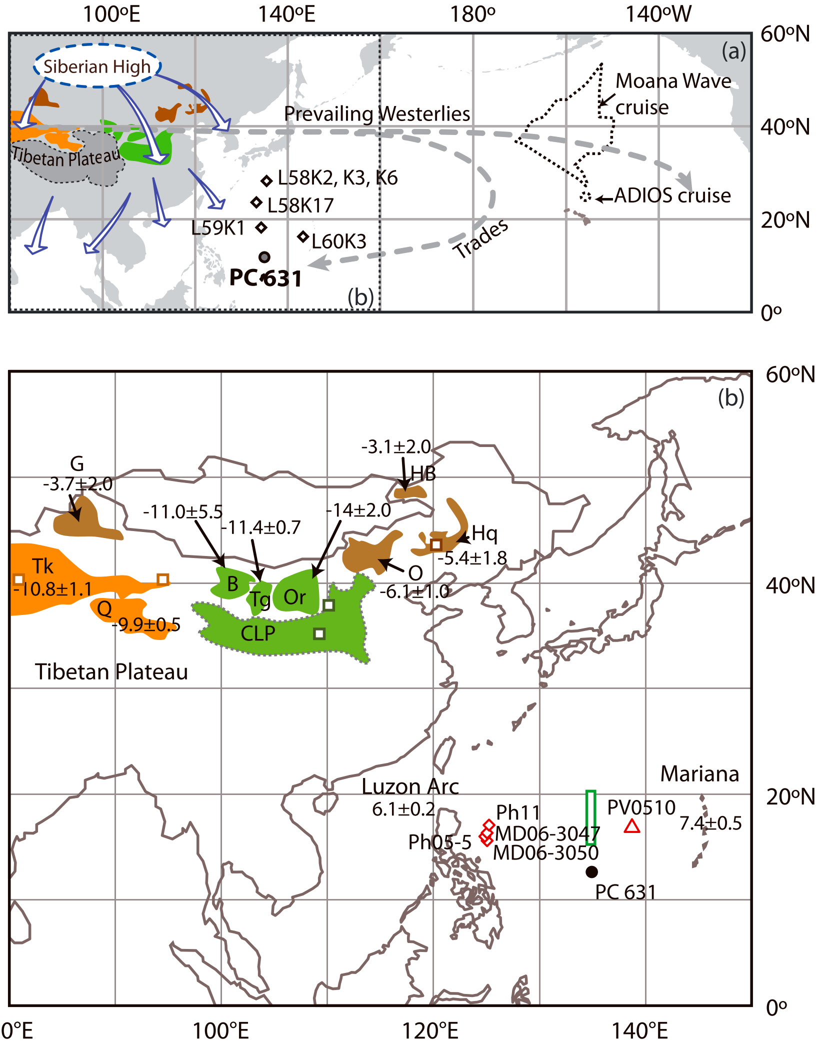

Figure 1. (a) Location of core PC 631 (solid gray circle, this study) and aerosol samples (open diamonds) collected during the *Moana Wave* cruise [*Arnold et al*., 1998], ADIOS cruise [*Arnold et al*., 1998], and a series of Deep Sea Drilling Project cruises (L series samples) [*Leinen et al*., 1994]. Also shown are the Asian winter monsoon wind directions and trajectory of the Prevailing Westerlies and Trades [*Merrill et al*., 1989; *Zhang et al*., 1997]. (b) Locations of sediment cores (open diamond: Ph11, Ph05-5, MD06-3047, and MD06-3050; open triangle: PV0510) and surface sediments (Parece Vela Basin, green rectangle). The stations where the aerosol dust samples were collected by *Shen et al*. [2005] are marked as open rectangles. Locations of possible dust source regions (i.e., Chinese deserts, Luzon Island, and the Mariana Islands) and their reported ε_{Nd} compositions are also shown [*Chen et al*., 2007; *Defant et al*., 1990; *Dixon and Stern*, 1983; *Honda et al*., 2004; *Nakano et al*., 2004; *Rao et al*., 2008; *Woodhead*, 1989]. The Asian deserts include the northern Chinese deserts (NCDs): the Gurbantunggut Desert (G), Onqin Daga Sandy Land (O), Hunlun Buir Sandy Land (HB), and Horqin Sandy Land (Hq); the Central Asian Deserts (CADs): the Taklimakan desert (Tk) and Qaidam Desert (Q); and the East Asian deserts (EADs): the Badain Jaran Desert (B), Tengger Desert (Tg), Ordos Desert (Or), and Chinese Loess Plateau (CLP). Modified from *Seo et al*. [2014].

evaluated because it is not possible to constrain relative contribution of CAD and BT dust. For this reason, *Seo et al*. [2014] used the ε_{Nd}-^{87}Sr/^{86}Sr composition only to estimate the relative contribution of Asian dust (~70%) and Luzon components (~30%) (Figure 2a), which is consistent with the values estimated from clay mineral composition (Figure 2b).

Xu et al. [2016] presented ε_{Nd} and ^{87}Sr/^{86}Sr compositions of three core sediments (Ph05-5, Ph11, and MD06-3047; see Figure 1b for locations) from the Benham Rise in a ε_{Nd}-^{87}Sr/^{86}Sr cross plot, in which these data plot in between two mixing lines drawn for Luzon-CAD/BT and Luzon-Ordos (Figure 2a). They suggest from this pattern that EAD has been a dominant (or at least strong) dust source over CAD in the Philippine Sea.

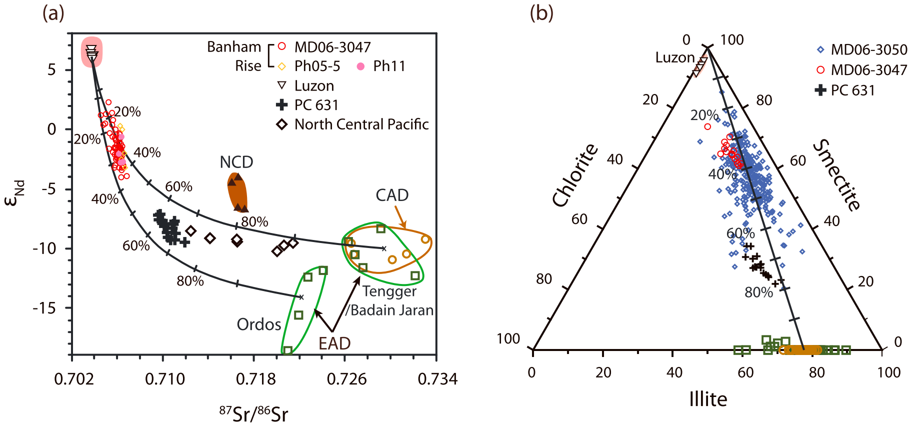

Figure 2. (a) ε_{Nd}-$^{87}Sr/^{86}Sr$ and (b) clay mineral composition of PC 631 (solid crosses), MD06-3047 (open red circles), Ph05-5 (open yellow diamonds), Ph11 (solid pink circles), and MD06-3050 (open blue diamonds) [*Jiang et al*., 2013; *Seo et al*., 2014; *Wan et al*., 2012; *Xu et al*., 2012, 2015]. Also shown are compositions of north central Pacific (NCP) surface sediment (diamonds), volcanic arcs in the western Pacific (inversed triangles), and soils in Chinese deserts grouped into three regions (NCD, CAD, and EAD refer to the northern Chinese deserts, Central Asian Deserts, and East Asian deserts, respectively).

However, their interpretation cannot be justified without quantification of CAD and BT contributions in theses sediments. Furthermore, the Benham Rise, only 250 km away from the Luzon Island, has been under the strong influence of Luzon component; 60–90% in ε_{Nd}-$^{87}Sr/^{86}Sr$ and 50–80% in clay mineral composition (Figure 2). As these data plot close to Luzon end-member composition on the mixing trend, it seems inappropriate to discuss the relative contribution of EAD and CAD dust with ε_{Nd} and $^{87}Sr/^{86}Sr$ compositions taking account of largely overlapping composition between EAD and CAD dust.

3. Clay Mineral Assemblages

There is a difficulty in quantifying the contribution of CAD and EAD dust based on the Nd-Sr isotope compositions owing to the common source rocks available to CADs and EADs [*Rao et al*., 2015] and long-range mobilization of sand dunes, from one to the other deserts, by eolian and fluvial processes through the geological history [*Nie et al*., 2015; *Sun et al*., 2008; *Zhao et al*., 2014]. On the other hand, clay mineral composition of a region reflects postdepositional weathering processes controlled by local climate. Especially, kaolinite is a product of intense chemical weathering [*Biscaye*, 1965; *Fagel*, 2007; *Griffin et al*., 1968] and thus its abundance helps to discriminate the region under the influence of the East Asian Summer Monsoon (e.g., EADs) from the hyperarid region (e.g., CADs). Indeed, *Shen et al*. [2005] reported a systematic eastward increase of kaolinite abundance in aerosols collected during EAWM season at five locations of Asian interior (Figure 1b for the locations). In a kaolinite/chlorite (K/C) and illite/kaolinite (I/K) cross plot, dust collected downwind of CADs and EADs forms distinctive fields with slight overlaps (Figure 3). Mineral composition of aerosols collected during the spring season in central and northwest Pacific (see Figure 1a for locations) plots in CAD and EAD fields in a K/C-I/K diagram, respectively, suggestive of mineral composition as a tool for source discrimination between CAD and EAD dust (Figure 3b). With an exception, ADIOS (Asian Dust Input to Oligotrophic Seas) aerosol data from the central Pacific plot in both EAD and CAD fields, which indicates a long-range transport of EAD dust to the central Pacific during the EAWM season [*Seo et al*., 2014]. K/C and I/K ratios have also been used successfully to characterize eolian dust in different climate settings and various locations [*Biscaye et al*., 1997; *Caquineau et al*., 1998; *Shen et al*., 2005]. These studies demonstrate usefulness of clay mineralogy as a source discriminator despite relatively large error (~±5%) associated with X-ray diffraction analysis.

The Luzon sediments collected in 21 rivers have been reported to consist mainly of smectite (86 ± 9%) with minor amounts of kaolinite (9 ± 8%), chlorite (5 ± 8%), and illite (1 ± 1%) ($n = 35$, ±1 SD) and do not show strong island-wide variations in composition [*Liu et al*., 2009]. Since Luzon sediments are enriched in kaolinite ($K/C = 1.8$ and $I/K = 0$ on average) compared to Asian dust ($K/C < 1$ and $I/K > 1$) [*Biscaye et al*., 1997; *Shen et al*., 2005] (Figure 3a), deep-sea sediments are expected to have higher K/C and lower I/K ratios with its increasing contribution of Luzon component. K/C and I/K ratios also vary with relative contributions of EAD and CAD

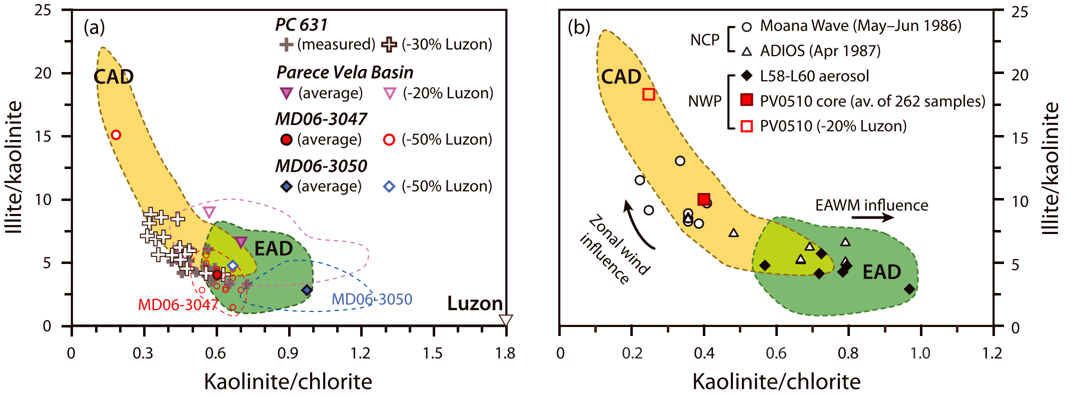

Figure 3. Kaolinite/chlorite-illite/kaolinite diagram for (a) PC 631 surface and downcore samples from the Parece Vela Basin [*Jin et al.*, 2007] and Benham Rise (MD06-3047 and MD06-3050) [*Wan et al.*, 2012; *Xu et al.*, 2012] and (b) for aerosol samples collected in the north central Pacific (NCP) (*Moana Wave* and ADIOS) and in the northwest Pacific (NWP) (L series) [*Arnold et al.*, 1998; *Leinen et al.*, 1994]. The average composition of 262 samples in a sediment core (PV0510) [*Ming et al.*, 2014] collected in the Philippine Sea is also plotted. Estimated compositions of Asian dust after removing the Luzon contribution are plotted as open symbols. Data fields for dust from the Central Asian Deserts (CAD) and East Asian deserts (EAD) are defined based on mineral composition of aerosols collected downwind of each source region [*Shen et al.*, 2005].

dust (Figure 3): thus, it is necessary to eliminate the mineralogical contribution of Luzon component to properly evaluate relative importance of EAD and CAD dust. To obtain K/C and I/K ratios of dust from Asian sources, we estimated the contribution of Luzon component from ε_{Nd}-$^{87}Sr/^{86}Sr$ and clay mineral composition (Figure 2) and deducted its mineralogical contribution to K/C and I/K values of bulk sample assuming simple mixing model. *Seo et al.* [2014] estimated 30% of Luzon component contribution based on ε_{Nd}-$^{87}Sr/^{86}Sr$ and clay mineral composition data, both of which indicated 30 to 40% of the Luzon component (Figure 2) and eliminate mineralogical contribution of Luzon component to decipher the mineralogical composition of Asian dust component. *Seo et al.* [2014] presented both uncorrected (bulk composition including Luzon contribution, black cross symbol) and corrected (excluding 30% of Luzon contribution, open cross symbol) mineral compositions, in which most of Asian dust end-member plot in the CAD dust field (Figure 3a). *Xu et al.* [2016] argue with uncorrected data [*Xu et al.*, 2016, Figure 3a] that PC631 sediments show close affinity to EAD dust, which we do not agree based on the discussion presented above. The clay mineralogy of a Philippine Sea sediment core, PV0510, recovered in close proximity to our study site (see Figure 1b for location) also plots in the CAD field [*Ming et al.*, 2014; *Seo et al.*, 2014] and so does it after subtracting Luzon component on the assumption of 20% of Luzon contribution (Figure 3b).

Xu et al. [2016] presented the clay mineral composition of sediments from various sites in the Philippine Sea (i.e., Banham Rise and Parece Vela Basin), all of which reveal clay mineral composition plotting closer to the EAD dust field. The sediments from the Benham rise (MD06-3047) include ~50–90% and 60–80% contribution of Luzon component in a ε_{Nd}-$^{87}Sr/^{86}Sr$ cross plot (Figure 2a) and clay mineral composition (Figure 2b), respectively [*Xu et al.*, 2012, 2015]. Based on this consistency, all the Benham Rise sediments (i.e., Ph05-5, Ph11, MD06-3047, and MD06-3050) [*Jiang et al.*, 2013; *Wan et al.*, 2012; *Xu et al.*, 2012, 2015] can be assumed to include minimal Luzon contribution of 50%. Parece Vela Basin sediments lacking ε_{Nd}-$^{87}Sr/^{86}Sr$ and raw clay mineral composition data can be assumed likely to include less than 30% of Luzon component considering its location about 500–800 km farther away from the Luzon source than to PC 631 (Figure 1b). When applying the correction for the contribution of Luzon component (50% for Benham Rise and 20% for Parece Vela Basin), the estimated average mineral compositions of Asian end-member plot in the CAD field (Figure 3a), suggesting that CAD dust dominates over EAD dust in the Philippine Sea. It is uncertain that such a correction is also applicable for the Banham Rise sediments taking account of the significant contribution of Luzon volcanic component reaching up to 90% and the compositional variation of Luzon sediment [*Liu et al.*, 2009]. However, this approach clearly demonstrates invalidity of using mineral composition data disregarding the contribution of Luzon component in evaluating relative importance of EAD and CAD dust.

Another issue raised by *Xu et al.* [2016] is the possibility for alteration of clay mineral during the long-range transport and by postdepositional processes. These issues have been of interest in clay mineralogy of deep-sea sediment and hence well studied. Previous works indicate that (i) there is no clear evidence for postdepositional alteration or authigenic formation of illite, kaolinite, and chlorite in the ocean interior [*Moriarty*, 1977], (ii) illite and kaolinite are resistant to chemical alteration in seawater [*Biscaye*, 1965; *Gradusov*, 1974; *Windom*, 1976], and (iii) authigenic chlorite is formed mostly by alteration of oceanic crust or contact metamorphism of sediments intruded by basaltic lavas [*Chamley*, 1989]. Thus, we preclude possibility of alteration by postdepositional process except for smectite that can be formed by alteration of volcanic or hydrothermal origin materials under water [*Chamley*, 1989]. Therefore, the deep-sea sediments of interest likely have preserved the I/K and K/C signatures of their sources.

4. Concluding Remarks

Against the conclusion of *Seo et al*. [2014], *Xu et al*. [2016] insisted that the Philippine Sea sediment shows closer resemblance to EAD than CAD dust in Nd-Sr isotopes and clay mineral compositions with the additional data. However, the relative contribution of CAD and EAD dust in the Philippine Sea cannot be resolved clearly with $\varepsilon_{\mathrm{Nd}}$ and $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$ compositions because of partial overlaps in composition of these two sources. Furthermore, the estimated contribution of Asian dust in the presented new data accounts only for 10–50%, which does not allow reliable assessment of relative contribution of EAD- and CAD-sourced dust. Also, the clay mineral composition presented by *Xu et al*. [2016] includes strong signals of Luzon soil that shifts I/K and K/C signatures toward the EAD dust field. With the removal of the Luzon signal, clay mineral composition of Asian end-member in these samples shows overall resemblance to CAD dust rather than EAD dust. Therefore, the additional data, presented by *Xu et al*. [2016], suggest significant contribution of EAD dust including Ordos Desert component but support the major conclusion of *Seo et al*. [2014] that CAD dust carried on the Prevailing Westerlies and Trade Winds dominates over EAD dust transported on EAWM winds in overall dust budget of the central Philippine Sea. Although this study was not able to address temporal changes in contribution of different source regions because of low temporal resolution of data, further study dealing with this aspect is recommended to better understand changes in dominant provenance and transportation mechanism of Asian dust in the past.

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