Calcite dust and the atmospheric supply of Nd to the Japan Sea

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Abstract

We report neodymium concentrations and isotope ratios (εNd) measured in the carbonate and silicate fractions of Chinese loess. The goal of the study is to investigate if the atmospheric supply of dissolved calcite dust to the oceans influences the cycling of Nd in seawater. We primarily focus on the Japan Sea, because it receives large quantities of dust from eastern Asia, and it has a known Nd isotope composition and a simple hydrologic budget. A steady-state atmospheric transport model is used to calculate the flux of soluble Nd delivered to the Japan Sea surface by the wet deposition of dissolved calcite dust. This information is then incorporated in a steady-state two-box model in order to evaluate controls on dissolved εNd values as well as to calculate the residence time of Nd in seawater.

On average, calcite in fresh Chinese loess contains ∼150 nmol of Nd g−1 with an εNd of −8.6. The atmospheric input of dissolved calcite to the Japan Sea surface is 7.4×1012 pmol yr−1, which corresponds to a dissolved Nd flux of 1.1×1018 pmol yr−1. This flux represents the largest input of Nd to the basin, and it is required to produce the εNd composition of Japan Sea waters. Approximately 65% of the Nd flowing from the Japan Sea surface into the western North Pacific originates directly from the input of dissolved calcite dust. Downwelling surface water and the remineralization of settling particles containing Nd scavenged from surface water contribute ∼77% of the Nd in the Japan Sea deep, which indicates that ∼50% of the deep Nd originates from calcite dust. The εNd composition of deep water is balanced by a benthic flux, but the input is a factor of ∼10 lower than the calcite dust flux and accounts for only ∼23% of the deep Nd. Surface and deep waters in the Japan Sea have Nd residence times, with respect to particle scavenging, of ∼14 and 530 yr, respectively. These findings reveal that the atmospheric supply of dissolved calcite dust to the Japan Sea is a more important component of the marine Nd cycle in this basin than previously recognized.

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1. Introduction

Because different water masses in the world’s oceans have different neodymium isotope ratios (143Nd/144Nd), Nd is widely utilized as a natural tracer of water mixing in and between various ocean basins [1–7]. However, current knowledge about the marine Nd cycle is incomplete. The 143Nd/144Nd ratios observed in seawater principally reflect the age and 147Sm/144Nd ratios of the continental rocks from which the Nd was derived (147Sm decays to 143Nd with T1/2 = 1.06×1011 yr) [7] and references therein. In order to explain these patterns, the residence time of Nd in seawater (τNd) must be less than the oceanic mixing time (∼103 yr). While it is generally agreed that τNd should be on the order of ∼2–
5×10^2 yr, dividing the total Nd inventory of seawater (∼4.4×10^{10} mol) by the global input from rivers (∼3.6×10^6 mol/yr) yields a value for τ_{Nd} that is significantly higher (∼10^4 yr) [2–6,8,9]. To resolve this problem, considerable effort has focused on identifying additional sources of Nd to seawater. Recent studies have proposed that dissolution/exchange processes involving coastal sediments, benthic fluxes, or the dissolution of atmospheric dust could provide enough Nd to lower calculations of τ_{Nd} to the expected level, but the issue remains equivocal [2,4,6,8–20].

The goal of the present work is to better understand how atmospheric processes influence the marine Nd cycle. In a previous study focusing on seawater Sr, Jacobson [21] identified the atmospheric supply of dissolved calcite dust from the Chinese Loess Plateau to the North Pacific as an important mechanism for delivering solutes to seawater. This research showed that calcite dust, which comprises only ∼10 wt.% of fresh loess, is rapidly dissolved during transport in the atmosphere. Because calcite dust originating from eastern Asia is known to contain relatively high concentrations of Nd [22], it follows that the wet deposition of dissolved calcite dust at the sea surface may contribute to the Nd inventory of the oceans.

To test this hypothesis, we primarily focus on the Japan Sea, because it receives large quantities of dust from eastern Asia [23]. The Japan Sea also has a simple hydrologic budget [24,25] and a known Nd isotope composition [3]. Thus, the system is an exceptional candidate for elucidating the role of atmospheric processes in the cycling of seawater Nd isotopes. First, we analyze the Nd geochemistry of both calcite and silicate dust collected from the Chinese Loess Plateau. Second, we use an atmospheric transport model to estimate the flux of Nd delivered to the Japan Sea by the dissolution of calcite dust. Third, we incorporate the atmospheric Nd flux into a two-box model of the Japan Sea in order to calculate the residence time of Nd in seawater. Our findings ultimately demonstrate that the flux of dissolved calcite to the Japan Sea yields a sizable input of Nd that is critical for residence time calculations.

2. Hydrology and Nd isotope geochemistry of the Japan Sea

The 1×10^6 km^2 Japan Sea is a marginal sea of the western North Pacific bounded to the west by Asia and to the east by the Japanese Islands (Fig. 1). Excluding the continental shelf area shallower than 200 m (∼10% of the total surface area), the mean depth of the basin is 1500 m [26]. The Tsushima Current carries warm, saline Kuroshio Water from the East China Sea into the Japan Sea through the Tsushima Strait at a rate of 4.7×10^{13} m^3 yr^{-1} [25]. The riverine input of fresh water to the basin is negligible (∼3.1×10^{11} m^3 yr^{-1}) [25]. Water exits the Japan Sea through the Tsugaru, Soya, and Mamiya straits to the north [24]. Within the basin, water is transported by the Tsushima Warm Current, the East Korean Warm Current, the Liman Current, and the North Korean Cold Current [24]. Based on the distribution of salinity, the Japan Sea surface (jss) is separated from the Japan Sea deep (jsd) at a depth of 200 m [26]. Water in the jsd is not formed by direct inflow from the East China Sea. Rather, the jss and jsd exchange water at a rate of 1.0×10^{13} m^3 yr^{-1} when surface water cools and sinks during winter [26].

Kuroshio water entering the Japan Sea contains 3.4×10^3 pmol m^{-3} of Nd with an ε_{Nd} value of −4.90 [2]. In sharp contrast, the jss contains between 2.2 and 3.1×10^4 pmol m^{-3} of Nd (avg.=2.4×10^4) with ε_{Nd} values ranging from −7.20 to −8.90 (avg.=−7.83) [3]. The ε_{Nd} values of the jss are distinct from those observed in the Okhotsk Sea and the western North Pacific, which supports the contention that reversal flow through the northern straits is relatively minor compared to inflow.
from the south [25]. The jsd contains \( \sim 4.5 \times 10^4 \) pmol m\(^{-3}\) of Nd [27] with an \( \varepsilon_{\text{Nd}} \) value of −7.00 [28].

3. Methods

We measured elemental concentrations and \( ^{143}\text{Nd}/^{144}\text{Nd} \) ratios in the carbonate and silicate fractions of 9 samples from the Chinese Loess Plateau. The samples are the same as those reported in [21]. To selectively dissolve the carbonate fraction of loess without leaching Nd from more resistant phases (e.g., chlorite, iron oxides, and silicates), we followed the procedure given in [29]. Accordingly, 1–3 g samples of uncrushed loess were mixed with ultrapure 5% acetic acid (AcOH) at 75 °C for 2 h. The procedure is similar to methodology presented in [20], where 10% acetic acid was used to selectively isolate the carbonate fraction of dust without dissolving other phases. The mixtures were centrifuged and the supernatant was collected. The remaining insoluble residue was rinsed several times with ultrapure water, dried, and weighed. Approximately 0.1 g subsamples of the dried residue were completely digested in ultrapure concentrated HF and HNO\(_3\) acids.

Ca and Fe concentrations in the leachates were measured by using an OPTIMA 4300 ICP-OES at the Saskatchewan Research Council in Saskatoon. The data have an uncertainty of ±10% (2\( \sigma \)). The calcite content of the samples was calculated by attributing all of the measured Ca to CaCO\(_3\) dissolution. Fe was measured to ensure that the leaching procedure did not dissolve Nd-rich, Fe-bearing phases [29].

Sm and Nd concentrations and isotope ratios were measured by using a Thermo Electron Triton (TIMS) in the Saskatchewan Isotope Laboratory at the University of Saskatchewan. Aliquots of the leachates and digests were equilibrated with a mixed \( ^{149}\text{Sm}−^{150}\text{Nd} \) spike, and Sm and Nd were separated by ion exchange chromatography following standard procedures. The purified fractions were loaded onto side filaments of double Re filament assemblies. An exponential law was used to correct for mass fractionation, and an iterative procedure was used to correct for the addition of the spike isotopes. The Sm isotope ratios were normalized to a \( ^{148}\text{Sm}/^{154}\text{Sm} \) ratio of 0.49419, and the \( ^{143}\text{Nd}/^{144}\text{Nd} \) ratios were normalized to a \( ^{146}\text{Nd}/^{144}\text{Nd} \) ratio of 0.7219. The uncertainty of the Sm and Nd concentrations is <1% (2\( \sigma \)). The \( ^{147}\text{Sm}/^{144}\text{Nd} \) ratios have an uncertainty of ±0.1% (2\( \sigma \)), and the \( ^{149}\text{Nd}/^{144}\text{Nd} \) ratios have an uncertainty of ±22 ppm (2\( \sigma \)). During the period of analysis, the measured \( ^{143}\text{Nd}/^{144}\text{Nd} \) ratio for the La Jolla standard was 0.511854. All Nd isotope data are reported in epsilon notation (\( \varepsilon_{\text{Nd}} \)), where \( \varepsilon_{\text{Nd}} = [(^{143}\text{Nd}/^{144}\text{Nd} − 0.512638)/0.512638] \times 10^4 \). In \( \varepsilon_{\text{Nd}} \) notation, the uncertainty is ±0.2 (2\( \sigma \)).

4. Results

Results for the leaching and digestion experiments are shown in Table 1. The Sm and Nd concentrations in the carbonate fraction of loess, here assumed to consist entirely of calcite dust, were determined by using the leachate Sm/Ca and Nd/Ca ratios, respectively. Because the Ca measurements by ICP-OES have a greater uncertainty than the Sm and Nd measurements by isotope dilution (10% versus <1%), we estimate that the Sm and Nd concentrations reported with respect to calcite have an uncertainty of 10%. The Sm concentrations range from 19 to 54 nmol g\(^{-1}\), and the Nd concentrations range from 94.3 to 253 nmol g\(^{-1}\). The average Sm and Nd concentrations are 32 and 152 nmol g\(^{-1}\), respectively. These results are directly comparable to previously reported data for calcite in Chinese loess and desert sands.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Concentrations and isotope ratios of Sm and Nd in the carbonate and silicate fractions of Chinese loess</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample</td>
<td>Sm(^\text{a}) (nmol g(^{-1}))</td>
</tr>
<tr>
<td>Binxian L1-1</td>
<td>29</td>
</tr>
<tr>
<td>Lantian L1-1</td>
<td>29</td>
</tr>
<tr>
<td>Lantian L1-4</td>
<td>54</td>
</tr>
<tr>
<td>Mubo L1-1</td>
<td>19</td>
</tr>
<tr>
<td>Mubo L1-4</td>
<td>28</td>
</tr>
<tr>
<td>Ningxian L1-1</td>
<td>32</td>
</tr>
<tr>
<td>Ningxian L1-4</td>
<td>27</td>
</tr>
<tr>
<td>Qingxian L1-1</td>
<td>35</td>
</tr>
<tr>
<td>Qingxian L1-4</td>
<td>37</td>
</tr>
</tbody>
</table>

\( \text{a} \) Per gram of CaCO\(_3\).

\( \text{b} \) Per gram of residue remaining after acetic acid leach.
Iron was not detected in any of the leachates, which suggests that Sm and Nd do not originate from the dissolution of Fe-bearing phases. We conclude from this observation that the leachate chemistry provides a reasonable estimate for the reservoir of Sm and Nd that will be released into natural waters when calcite dust dissolves.

The carbonate fraction of loess has $^{147}\text{Sm}/^{144}\text{Nd}$ ratios and $\varepsilon_{\text{Nd}}$ values ranging from 0.1311 to 0.1362 and from $-7.7$ to $-8.9$, respectively. The average values are 0.1340 and $-8.6$, respectively. To our knowledge, no studies have reported $^{147}\text{Sm}/^{144}\text{Nd}$ ratios of the carbonate fraction in Chinese loess, and only one study has reported $\varepsilon_{\text{Nd}}$ values ranging from $-9.1$ to $-10.7$ ($\pm 0.5$, 2$\sigma$) [20]. These latter data are slightly less radiogenic than the results presented in Table 1, but given the reported uncertainties in this study and the study of [20], some overlap is evident.

The Sm and Nd concentrations in the silicate fraction of loess are reported per gram of insoluble residue. These concentrations range from 37 to 44 and from 199 to 237 nmol g$^{-1}$, respectively. The average Sm concentration is 40 nmol g$^{-1}$, and the average Nd concentration is 215 nmol g$^{-1}$. The $^{147}\text{Sm}/^{144}\text{Nd}$ ratios and $\varepsilon_{\text{Nd}}$ values range from 0.1161 to 0.1191 and from $-9.80$ to $-11.1$, respectively. The average values are 0.1176 and $-10.2$, respectively. With these data, we find that the silicate fraction has an average Nd model age of 1.5 Ga. The concentration, isotope, and model age findings reported here are directly comparable to results reported elsewhere [20,30–32].

5. Discussion

A general analysis of the Japan Sea Nd budget presented in Section 2 provides insight into the nature of the sources required to produce the observed water data. The dramatic compositional change in surface water chemistry across the Tsushima Strait cannot be attributed to riverine inflow, because the Nd concentration in rivers is generally low [33] and the freshwater input to the Japan Sea is negligible [25]. Likewise, the compositional change is not due to mixing between inflowing Kuroshio water and upwelling deep water. Neodymium from the East China Sea ($\varepsilon_{\text{Nd}} = -4.9$) flows into the JSS at a rate of $1.6 \times 10^{17}$ pmol Nd yr$^{-1}$, while Nd from the JSD ($\varepsilon_{\text{Nd}} = -7.0$) upwells into the JSS at a rate of $4.5 \times 10^{17}$ pmol Nd yr$^{-1}$. The isotopic mass balance between these inputs is $-6.45$, which is more radiogenic than the average observed value of $-7.83$. Thus, it is clear that an additional Nd input to the Japan Sea is required and that the input must be comparatively large, relatively unradiogenic, and delivered directly to the surface of the Japan Sea.

5.1. Calcite dust atmospheric transport model

5.1.1. Model description

Because the isotope composition of calcite dust is the right magnitude ($-8.6$) to balance mixing between the Kuroshio and upwelling inflows to the JSS, we propose that the dissolution of calcite dust emanating from eastern Asia is important for the Japan Sea Nd budget. To constrain the Nd flux from this source, it is necessary to quantify the rate at which dissolved calcite dust is transferred from the atmosphere to the Japan Sea surface by wet deposition. The model described below roughly follows the approach used to examine the transport of $^{210}\text{Pb}$ over the North Pacific [34]. Accordingly, we assume that 1) Dust is injected into the atmosphere at the Asian coastline; 2) transport occurs along the prevailing wind direction at a constant velocity; 3) the effect of dispersion is negligible; and 4) all reaction rates are first-order. We also assume that calcite dust dissolves during interaction with water vapor [35] and that the resulting aerosols are scavenged from the atmosphere by rainwater falling into seawater. We do not distinguish between in-cloud and below-cloud scavenging. Also, we do not consider the dissolution of solid calcite dust delivered to the Japan Sea, because the rate of calcite dissolution in seawater is much slower than that in the atmosphere [21]. As demonstrated below, our deposition estimates for dissolved calcite dust, as well as the remaining insoluble fraction of dust, are in very good agreement with both measured and modeled data from other studies.

We consider the transport of both solid and dissolved calcite dust ($\text{scal}$ and $\text{dcal}$, respectively). Solid calcite dust is advected by wind and removed by dry deposition, wet deposition, and dissolution. Calcite in the dissolved phase is advected by wind, added by dissolution of solid calcite dust, and removed by wet deposition. The general transport equations for $\text{scal}$ and $\text{dcal}$, respectively, are:

$$\frac{dC_{\text{scal}}}{dt} = -v \frac{dC_{\text{scal}}}{dx} (\lambda_{\text{dry}} + \lambda_{\text{wet}} + \lambda_{\text{cal}}) C_{\text{atm}}$$

$$\frac{dC_{\text{dcal}}}{dt} = -v \frac{dC_{\text{dcal}}}{dx} + \lambda_{\text{cal}} C_{\text{atm}}$$

where $C_{\text{scal}}$ and $C_{\text{dcal}}$ are the concentrations of solid and dissolved calcite dust in a vertical well-mixed column of the atmosphere, respectively; $v$ is the wind velocity; $x$ is the transport distance; $\lambda_{\text{dry}}$ is the dry deposition rate;
\( z_{\text{scal}} \) is the wet deposition rate of scal; \( z_{\text{dcal}} \) is the wet deposition rate of dcal; and \( \lambda_{\text{cal}} \) is the dissolution rate of calcite. The wet deposition rates for scal and dcal are differentiated, because wet deposition rates reported for solid particles are typically different than those reported for dissolved ions [23,36–40].

The rate constants in Eqs. (1) and (2) are calculated according to the following equations:

\[
\begin{align*}
\lambda_{\text{dry}} &= v_{\text{dry}}/h_{\text{atm}} \\
\lambda_{\text{wet}} &= PS_i \rho_w/\rho_a h_{\text{atm}} \\
\lambda_{\text{cal}} &= \omega_{\text{cal}} \sigma_{\text{cal}}
\end{align*}
\]

where \( v_{\text{dry}} \) is the dry deposition velocity of dust particles, \( h_{\text{atm}} \) is the height over which dust is distributed in the atmosphere, \( P \) is the rainfall rate, \( S_i \) is the scavenging rate ( \( i = \text{scal} \) or dcal), which is the dimensionless concentration ratio of dust in rainwater versus that in the atmosphere, \( \rho_w \) is the density of water, \( \rho_a \) is the density of air, \( \omega_{\text{cal}} \) is the surface area normalized calcite dissolution rate, and \( \sigma_{\text{cal}} \) is the specific surface area of calcite dust.

The steady-state solutions to Eqs. (1) and (2) yield the following expressions for the concentrations of solid and dissolved calcite dust in the atmosphere as a function of transport distance [\( C_{\text{atm}}^{\text{scal}}(x) \) and \( C_{\text{atm}}^{\text{dcal}}(x) \), respectively]:

\[
\begin{align*}
C_{\text{atm}}^{\text{scal}}(x) &= C_{\text{atm}}^{\text{scal}}(o) \exp \left[ -\frac{x}{v} (\lambda_{\text{dry}} + \lambda_{\text{wet}} + \lambda_{\text{cal}}) \right] \\
C_{\text{atm}}^{\text{dcal}}(x) &= \left[ C_{\text{atm}}^{\text{dcal}}(o) + \frac{\lambda_{\text{cal}} C_{\text{atm}}^{\text{cal}}(o)}{\lambda_{\text{cal}} + \lambda_{\text{dry}} + \lambda_{\text{wet}} - \lambda_{\text{cal}}} \right] \times \exp \left[ -\frac{x}{v} (\lambda_{\text{dcal}}) \right] - \frac{\lambda_{\text{cal}} C_{\text{atm}}^{\text{cal}}(x)}{\lambda_{\text{cal}} + \lambda_{\text{dry}} + \lambda_{\text{wet}} - \lambda_{\text{cal}}}
\end{align*}
\]

where \( C_{\text{atm}}^{\text{scal}}(o) \) and \( C_{\text{atm}}^{\text{dcal}}(o) \) are the initial concentrations of solid and dissolved calcite dust, respectively, in a vertical column of the atmosphere at the western coastline of the Japan Sea. \( C_{\text{atm}}^{\text{scal}}(o) \) is given by the equation \( C_{\text{atm}}^{\text{scal}}(o) = C_{\text{dust}}^{\text{cal}} f_{\text{cal}} h_{\text{atm}} \), where \( C_{\text{dust}}^{\text{cal}}(o) \) is the mean annual concentration of bulk dust in a vertical column of air at the coastline; \( f_{\text{cal}} \) is the mass fraction of calcite in bulk dust; and \( h_{\text{atm}} \) is defined above. \( C_{\text{atm}}^{\text{dcal}}(o) \) is given by the equation \( C_{\text{atm}}^{\text{dcal}}(o) = C_{\text{rain}}^{\text{dcal}}(o) \rho_w h_{\text{atm}} / S_{\text{atm}} P_{\text{atm}} \), where \( C_{\text{rain}}^{\text{dcal}}(o) \) is the mass of dissolved calcite per volume of rainwater and the remainder of the parameters are defined above.

The deposition rate of dissolved calcite dust at the surface of the Japan Sea as a function of transport distance [\( I_{\text{dcal}}^{\text{atm}}(x) \)] is,

\[
I_{\text{dcal}}^{\text{atm}}(x) = \lambda_{\text{wet}} C_{\text{atm}}^{\text{dcal}}(x)
\]

and the average deposition rate over the entire trajectory [\( I_{\text{dcal}}^{\text{atm}} \)] is:

\[
I_{\text{dcal}}^{\text{atm}} = \frac{1}{x_f-x_o} \int_{x_o}^{x_f} I_{\text{dcal}}^{\text{atm}}(x)dx
\]

where \( x_f \) and \( x_o \) are the initial and final positions along the transport path, respectively. With an estimate for \( I_{\text{dcal}}^{\text{atm}} \), it is then possible to calculate the flux of dissolved calcite \( (Q_{\text{dcal} \rightarrow \text{jss}}) \) and Nd \( (F_{\text{dcal} \rightarrow \text{jss}}) \) to the Japan Sea using the following equations:

\[
\begin{align*}
Q_{\text{dcal} \rightarrow \text{jss}} &= I_{\text{dcal}}^{\text{atm}} A_{\text{jss}} \\
F_{\text{dcal} \rightarrow \text{jss}} &= C_{\text{cal}}^{\text{Nd}} Q_{\text{dcal} \rightarrow \text{jss}}
\end{align*}
\]

where \( A_{\text{jss}} \) is the surface area of the Japan Sea, and \( C_{\text{cal}}^{\text{Nd}} \) is the Nd concentration in calcite dust.

Model input parameters discussed below are summarized in Table 2. Our goal is to conservatively estimate \( F_{\text{dcal} \rightarrow \text{jss}} \). Therefore, when possible, we use input values that describe mean annual conditions for dust transport over the Japan Sea. We use 1000 km for the width of the Japan Sea (\( x_i \)) and 5 m s\(^{-1} \) for the average wind velocity (v) [23,34]. Chinese loess, which contains \( \approx 10 \) wt.\% calcite [21], is commonly lofted to altitudes between 2000 and 6000 m [23] at a mean annual concentration of \( \approx 35 \) mg m\(^{-3} \) [23,41]. Hence, we use 0.1 for \( f_{\text{cal}} \), 4000 m for \( h_{\text{atm}} \), and 3.5 \( \times \) 10\(^{-5} \) g m\(^{-3} \) for \( C_{\text{dust}}^{\text{cal}}(o) \).

To calculate \( \lambda_{\text{dcal}} \), we use 0.005 m s\(^{-1} \) for \( v_{\text{dry}} \) [23,42]. Calculation of \( \lambda_{\text{wet}} \) requires knowledge of P and \( S_i \). We use 3.8 \( \times \) 10\(^{-8} \) m s\(^{-1} \) (\( \approx 1.2 \) m yr\(^{-1} \)) for P [43–45]. Estimates for \( S_i \) vary widely according to the size and solubility of the particle, compound, or charged ion being examined and the location of rainfall (i.e., continental versus marine) [23,36–40]. Recent studies indicate that \( S \) equals 200 for solid dust particles deposited at the sea surface [23,38]. By contrast, \( S \) equals 2100 for Ca\(^{2+} \) over the oceans [39]. Thus, we use 200 for \( S_{\text{cal}} \) and 2100 for \( S_{\text{dcal}} \). To calculate \( \lambda_{\text{cal}} \), we use 0.785 g cm\(^{-2} \) yr\(^{-1} \) for \( \omega_{\text{cal}} \) and 1000 cm\(^2 \) g\(^{-1} \) for \( \sigma_{\text{cal}} \) [35].

The value for \( \omega_{\text{cal}} \) corresponds to the dissolution rate of calcite at pH = 5.5 [46], which is roughly the pH of pristine rainwater in equilibrium with atmospheric CO\(_2 \) [47]. We consider this to be a reasonable estimate for \( \omega_{\text{cal}} \), as the mean annual pH of rainwater over eastern Asia and Japan ranges from \( \sim 4 \) to 7 [48,49].
Calculation of $C_{\text{atm}}(o)$ requires the values for $f_{\text{cal}}$, $h_{\text{atm}}$, and $C_{\text{atm}}(o)$ given above. This parameter equals $1.4 \times 10^{-2}$ g m$^{-2}$. To estimate the initial concentration of dissolved calcite dust in the atmosphere [$C_{\text{atm}}(o)$], we note that the concentration of non-sea-salt-Ca$^{2+}$ (nss-Ca$^{2+}$) in rainwater over northern China, weighted by mean annual rainfall, is $\sim 160$ mmol m$^{-3}$ [43-45]. By attributing nss-Ca$^{2+}$ in rainwater to the dissolution of calcite dust, which is well-known to be the case in this region [35,48-51], we estimate that the initial concentration of dissolved calcite dust in rainwater is $\sim 16$ g m$^{-3}$. By comparison, the theoretical concentration of dissolved calcite in equilibrium with pure water under standard temperature and pressure conditions is roughly a factor of $\sim 3$ higher [47]. Thus, our assumption about the origin of nss-Ca$^{2+}$ appears reasonable. Multiplying 16 g m$^{-3}$ by the quantity $\rho_{\text{atm}}/S_{\text{cal}}\rho_{\text{w}}$ yields $3.7 \times 10^{-2}$ g m$^{-2}$ for $C_{\text{atm}}(o)$.

Table 2: Parameters for the atmospheric transport model

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Description</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>$x$</td>
<td>Distance along transport path</td>
<td>m</td>
</tr>
<tr>
<td>$v$</td>
<td>Wind velocity</td>
<td>5 m s$^{-1}$</td>
</tr>
<tr>
<td>$h_{\text{atm}}$</td>
<td>Height of dust injection into the atmosphere</td>
<td>4$\times$10$^7$ m</td>
</tr>
<tr>
<td>$f_{\text{cal}}$</td>
<td>Mass fraction of calcite in bulk dust</td>
<td>0.1</td>
</tr>
<tr>
<td>$A_{\text{jss}}$</td>
<td>Surface area of the Japan Sea</td>
<td>$1\times10^{12}$ m$^2$</td>
</tr>
<tr>
<td>$C_{\text{atm}}(o)$</td>
<td>Atmospheric concentration of bulk dust at $x=0$</td>
<td>$3.5 \times 10^{-2}$ g m$^{-3}$</td>
</tr>
<tr>
<td>$C_{\text{cal}}(o)$</td>
<td>Atmospheric concentration of solid calcite dust (cal) over $h_{\text{atm}}$ at $x=0$</td>
<td>$1.4 \times 10^{-2}$ g m$^{-2}$</td>
</tr>
<tr>
<td>$C_{\text{cal}}(o)$</td>
<td>Atmospheric concentration of dissolved calcite dust (dcal) over $h_{\text{atm}}$ at $x=0$</td>
<td>$3.7 \times 10^{-2}$ g m$^{-2}$</td>
</tr>
<tr>
<td>$C_{\text{Nd}}$</td>
<td>Concentration of Nd in calcite dust</td>
<td>$1.5 \times 10^7$ pmol g$^{-1}$</td>
</tr>
</tbody>
</table>

5.1.2. Atmospheric transport model results

Fig. 2a and b display the model output, and Table 2 summarizes the key results. From west to east across the Japan Sea (i.e., from $x_o=0$ to $x_f=1000$ km), we calculate that $C_{\text{atm}}(x)$ decreases from $\sim 37$ to $2.2 \times 10^{-3}$ g m$^{-2}$ and that $f_{\text{dcal}}(x)$ decreases from $\sim 20$ to 1.0 g m$^{-2}$ yr$^{-1}$. From Eq. (9), we calculate that the average deposition rate of dissolved calcite dust at the Japan Sea surface ($F_{\text{dcal}}(\text{jss})$) is $\sim 7.4$ g m$^{-2}$ yr$^{-1}$. This corresponds to a dissolved calcite flux ($Q_{\text{dcal} \rightarrow \text{jss}}$) of $\sim 7.4 \times 10^{12}$ g yr$^{-1}$ and a Nd flux ($F_{\text{Nd}}(\text{dcal} \rightarrow \text{jss})$) of $\sim 1.1 \times 10^{10}$ pmol yr$^{-1}$. 

Immediate comparisons to $F_{\text{dcal}}(\text{atm})$, $Q_{\text{dcal} \rightarrow \text{jss}}$, and $F_{\text{Nd}}(\text{dcal} \rightarrow \text{jss})$ are not available in the literature, because no studies have reported deposition rates of dissolved calcite dust to the Japan Sea. However, data are available for the extremes of the trajectory (i.e., China and Japan). In these locations, the wet deposition rates of nss-Ca$^{2+}$ range from $\sim 80$ to 240 and from $\sim 3$ to 17 mmol m$^{-2}$ yr$^{-1}$, respectively [43-45]. Consistent with our previous assumption about the origin of nss-Ca$^{2+}$, we estimate that the deposition rates of dissolved calcite range from $\sim 8$ to 24 g m$^{-2}$ yr$^{-1}$ over China and from $\sim 0.3$ to 1.7 g m$^{-2}$ yr$^{-1}$ over Japan. These observations are in acceptable agreement with our predicted values of $C_{\text{atm}}(x)$ presented above. We can also check the model by calculating the average Nd concentration in rainwater [$C_{\text{rain}}(x)$]. These data are calculated with the equation
\( C_{\text{rain}}^{\text{Nd}} = I_{\text{atm}}^{\text{Nd}} / P \), where \( C_{\text{rain}}^{\text{Nd}} \) has units of pmol m\(^{-3}\). The model predicts that \( C_{\text{rain}}^{\text{Nd}} \) equals \( \sim 1 \times 10^5 \) pmol m\(^{-3}\) over Japan, which is in broad accord with the average measured value of \( \sim 8 \times 10^4 \) pmol m\(^{-3}\) [52].

An additional constraint on the model quality is given by adapting the equations to evaluate the fate and transport of insoluble mineral particles, here assumed to be silicate dust (sil). In this case, only modified versions of Eqs. (4), (6), and (8) are required. The changes are as follows: 1) To calculate the wet deposition rate of silicate dust (\( \lambda_{\text{wet}}^{\text{sil}} \)), we use \( S_{\text{sil}} = 200 \) in Eq. (4) [23]; 2) To calculate the concentration of silicate dust in the atmosphere \( C_{\text{atm}}^{\text{sil}}(x) \) from Eq. (6), we omit a dissolution term, because silicate mineral dissolution rates are typically 3 to 5 orders of magnitude lower than the dissolution rate of calcite; 3) To calculate the initial concentration of silicate dust in the atmosphere at the Japan Sea coastline \( C_{\text{atm}}^{\text{sil}}(0) \), we use the fraction of silicate dust \( f_{\text{sil}} \), where \( f_{\text{sil}} = 1 - f_{\text{cal}} \); 4) To calculate the deposition rate of silicate dust as a function of transport distance \( I_{\text{atm}}^{\text{sil}}(x) \), we replace \( \lambda_{\text{wet}}^{\text{dcal}} \) in Eq. (8) with the quantity \( \lambda_{\text{dry}} + 3 \lambda_{\text{wet}}^{\text{sil}} \). The average deposition rate of silicate dust \( (I_{\text{atm}}^{\text{sil}}) \) follows from Eq. (9).

The results for silicate dust are displayed in Fig. 2a and b. From west to east across the Japan Sea, we calculate that \( C_{\text{atm}}^{\text{sil}}(x) \) decreases from 1.3 to \( 0.71 \times 10^{-4} \) g m\(^{-2}\) and that \( I_{\text{atm}}^{\text{sil}}(x) \) decreases from 11 to 6.4 g m\(^{-2}\) yr\(^{-1}\). From Eq. (9), we estimate that \( I_{\text{atm}}^{\text{sil}} \) equals 8.6 g m\(^{-2}\) yr\(^{-1}\). This result agrees very well with the recently reported value of 8.2 g m\(^{-2}\) yr\(^{-1}\) [23]. From Eq. (10), the total flux of silicate dust to the Japan Sea is \( 8.6 \times 10^{12} \) g yr\(^{-1}\). Laboratory experiments have shown that \( \sim 3\% \) of the Nd contained in silicate dust is mobilized into seawater following deposition [13]. The average concentration of Nd in silicate dust is 215 nmol g\(^{-1}\) (Table 1), which indicates that the partial dissolution of silicate dust could supply \( \sim 5.5 \times 10^{16} \) pmol yr\(^{-1}\) to the Japan Sea. Because this Nd input is about two orders of magnitude less than the input from dissolved calcite dust, we do not consider the silicate dust input in subsequent calculations.
Overall, because our modeled values for dust deposition and rainwater chemistry are in general accord with previously reported data from a variety of studies, we conclude that the input parameters accurately reflect controls on dust transport and that our estimate of $F_{\text{dcal} \rightarrow \text{jss}}$ required for the oceanic box model below is reasonable. A final observation of note is that the rate of dissolved calcite deposition ($\bar{\gamma}_{\text{atm}}$) is not simply equal to $f_{\text{cal}} \bar{\gamma}_{\text{atm}}$, particularly close to the dust source region. This would lead to a significant underestimation of $F_{\text{dcal} \rightarrow \text{jss}}$. Although both calcite and silicate dust particles are likely emitted from arid regions in eastern Asia at the same rate, the processes governing their fate and transport are vastly different. The removal rate of dissolved calcite dust from the atmosphere is much faster than that of silicate dust, primarily because charged ions are more effectively scavenged from the atmosphere by rainwater than insoluble silicate particles. The net result is that the half-decrease distance for dissolved calcite dust is roughly a factor of 10 less than the half-decrease distance for insoluble silicate dust. Thus, the ratio of dissolved calcite to silicate dust in the atmosphere decreases with increasing distance away from the source region, and the relative deposition rates of calcite and silicate dust do not necessarily reflect their initial abundances prior to entrainment.

5.2. Two-box model of the Japan Sea Nd cycle

5.2.1. Model description

To evaluate how $F_{\text{dcal} \rightarrow \text{jss}}$ calculated above influences the residence time of Nd in the Japan Sea, we use a two-box model (Fig. 3a and b). All model parameters discussed below are summarized in Table 3. Consistent with the hydrologic description presented above, we assume that the Japan Sea is composed of two uniform reservoirs, the jss and the jsd, separated at a depth of 200m. The reservoir volumes are $V_{\text{jss}}$ and $V_{\text{jsd}}$. Each reservoir is characterized by a dissolved Nd concentration ($C_{\text{jss}}^{\text{Nd}}$ and $C_{\text{jsd}}^{\text{Nd}}$), a dissolved Nd isotope composition ($\varepsilon_{\text{Nd(jss)}}$ and $\varepsilon_{\text{Nd(jsd)}}$), and incoming and outgoing Nd fluxes ($F_{i \rightarrow j}^{\text{Nd}}$) with a characteristic isotope composition ($\varepsilon_{\text{Nd(i)}}$). Thus, advective fluxes are calculated according to the equation $F_{i \rightarrow j}^{\text{Nd}} = C_{i}^{\text{Nd}} Q_{i \rightarrow j}$, where $F_{i \rightarrow j}^{\text{Nd}}$ equals the amount of Nd transferred from source $i$ to sink $j$ per unit time, $C_{i}^{\text{Nd}}$ is the concentration of Nd in source $i$, and $Q_{i \rightarrow j}$ is the amount of source $i$ transferred to sink $j$ per unit time. Diffusive fluxes are calculated according to the equation $F_{i \rightarrow j}^{\text{Nd}}=(dC/dx)_{i}^{\text{Nd}} D_{\text{Nd}i} A_{\text{i}}$, where $(dC/dx)_{i}^{\text{Nd}}$ is
the concentration gradient of Nd in source $i$, $D_{Nd}$ is the Nd diffusion coefficient, and $A$ is the area.

At steady-state, the Nd budget and isotopic composition of the jss are expressed by the following equations, respectively:

$$
C_{Nd}^{Ku}Q_{Ku\rightarrow jss} + C_{Nd}^{Jsd}Q_{Jsd\rightarrow jss} + C_{cal}^{Nd}Q_{cal\rightarrow jss} = C_{jss}^{Nd}Q_{jss\rightarrow jsd} + C_{jss}^{Nd}Q_{jss\rightarrow wnp} + k_{jss}^{P}C_{jss}^{Nd}V_{jss}
$$

(12)

$$
epsilon_{Nd(jss)}C_{jss}^{Nd}Q_{jss\rightarrow jsd} + epsilon_{Nd(jsd)}C_{jss}^{Nd}Q_{jss\rightarrow jss} + eepsilon_{Nd(cal)}C_{cal}^{Nd}Q_{cal\rightarrow jss} = epsilon_{Nd(jss)}(C_{jss}^{Nd}Q_{jss\rightarrow jsd} + C_{jss}^{Nd}Q_{jss\rightarrow wnp} + k_{jss}^{P}C_{jss}^{Nd}V_{jss})
$$

(13)

where $C_{Nd}^{Ku}$ and $eepsilon_{Nd(ku)}$ are the concentration and isotopic composition of Nd in Kuroshio water, respectively; $Q_{Ku\rightarrow jss}$ is the flux of Kuroshio water to the jss through the Tsushima Strait; $Q_{Jsd\rightarrow jss}$ is the upwelling flux of jss water to the jss; $Q_{jss\rightarrow jsd}$ is the downwelling flux of jss water to the jsd; $Q_{jss\rightarrow wnp}$ is the flux of jss water to the western North Pacific through the northern straits of the Japan Sea; $C_{cal}$ and $eepsilon_{Nd(cal)}$ are the concentration and isotopic composition of Nd in calcite dust, respectively.

$Q_{cal\rightarrow jss}$ is the atmospheric flux of dissolved calcite dust to the jss calculated from Eq. (10); and $k_{jss}^{P}$ is the first-order rate constant for the scavenging of dissolved Nd from jss water by settling particles.

At steady-state, the Nd budget and isotopic composition of the jsd are expressed by the following equations, respectively:

$$
C_{jsd}^{Nd}Q_{jsd\rightarrow jss} + \beta k_{jsd}^{P}C_{jsd}^{Nd}V_{jsd} + (dC/dV)_{ben}^{Nd}D_{Nd}A_{jsd} = C_{jsd}^{Nd}Q_{jsd\rightarrow jss} + k_{jsd}^{P}C_{jsd}^{Nd}V_{jsd}
$$

(14)

$$
epsilon_{Nd(jsd)}(C_{jsd}^{Nd}Q_{jsd\rightarrow jss} + \beta k_{jsd}^{P}C_{jsd}^{Nd}V_{jsd}) + eepsilon_{Nd(ben)}(dC/dV)_{ben}^{Nd}D_{Nd}A_{jsd} = eepsilon_{Nd(jsd)}(C_{jsd}^{Nd}Q_{jsd\rightarrow jss} + k_{jsd}^{P}C_{jsd}^{Nd}V_{jsd})
$$

(15)

where $\beta$ is the fraction of settling particles from the jss that are remineralized in the jsd ($0 \leq \beta \leq 1$); $k_{jsd}^{P}$ is the flux of Nd to the jsd resulting from particle remineralization; $(dC/dV)_{ben}^{Nd}$ and $eepsilon_{Nd(ben)}$ are the concentration gradient and isotopic composition of Nd in benthic porewaters, respectively; $A_{jsd}$ is the surface area.
of the jsd (=A_jsd); and $k_{jsd}^p$ is the first-order rate constant for the scavenging of dissolved Nd from jsd water by settling particles.

The total residence times of Nd in the jss and jsd ($\tau_{jss}$ and $\tau_{jsd}$, respectively) reflect the combined effects of water flow through the reservoirs and particle scavenging. Analysis of Eqs. (12) and (14) demonstrates that the equations for $\tau_{jss}$ and $\tau_{jsd}$, respectively, are:

$$\tau_{jss} = \frac{C_{Nd}^j V_{jss}}{F_{jss-jsd} + F_{jss-wmp} + k_{jss}^p C_{jss}^p V_{jss}} = \left(\frac{1}{\tau_{jss}^H} + \frac{1}{\tau_{jss}}\right)^{-1}$$

(16)

$$\tau_{jsd} = \frac{C_{Nd}^j V_{jsd}}{F_{jsd-jss} + k_{jsd}^p C_{jsd}^p V_{jsd}} = \left(\frac{1}{\tau_{jsd}^H} + \frac{1}{\tau_{jsd}}\right)^{-1}$$

(17)

where the hydraulic retention time of water in the jss ($\tau_{jss}^H$) equals $V_{jss}/(Q_{jss-jsd} + Q_{jss-wmp})$; the residence time with respect to particle scavenging in the jss ($\tau_{jss}^d$) equals $1/k_{jss}^p$; the hydraulic retention time of water in the jsd ($\tau_{jsd}^H$) equals $V_{jsd}/Q_{jsd-jss}$; and the residence time with respect to particle scavenging in the jsd ($\tau_{jsd}^d$) equals $1/k_{jsd}^p$.

5.2.2. Ocean box model results

Results from the two-box ocean model are presented in Table 3. Determining $\tau_{jss}^Nd$ is relatively straightforward, as the calculation simply relies on the hydrologic balance of the jss, the measured Nd concentrations of the various water masses, and the value of $F_{dcal→jss}$ calculated above. By rearranging Eq. (12), we find that $k_{jss}^p$ equals $7.1 \times 10^{-2}$ yr$^{-1}$. Thus, the particle scavenging flux of Nd ($k_{jss}^p C_{jss}^p V_{jss}$) equals $3.4 \times 10^{17}$ pmol yr$^{-1}$. We can check the value of $k_{jss}^p$ by solving for $\varepsilon_{Nd(jss)}$ in Eq. (13). We calculate that $\varepsilon_{Nd(jss)} = -7.83$, which exactly matches the average value reported for the jss [3]. The residence time of Nd in jss water with respect to particle scavenging ($\tau_{jss}^d$) is $\sim 14$ yr. This value is broadly consistent with the residence times of 1.5–2.6 yr reported for a 100 m surface layer of the global ocean [2]. Because $\tau_{jss}$ is slightly shorter at $\sim 4$ yr, the jss should be reasonably well-mixed with respect to Nd. According to Eq. (16), we calculate that the total residence time of Nd in the jss reservoir ($\tau_{jss}^Nd$) equals $\sim 2.8$ yr.

Determining $\tau_{jsd}^Nd$ from Eq. (17) requires an estimate of $k_{jsd}^p$ from Eq. (14), which in turn requires the preceding calculation of $k_{jss}^p$ as well as knowledge about $\beta$ and $F_{Nd}^j$. To constrain $\beta$, we use data for the compositional change of settling particles in the Japan Sea [53]. Between depths of 890 and 3240 m, the latter of which is roughly the depth of the sediment-water interface in the deepest region of the Japan Sea, the total flux of settling organic matter, opal, calcite, and clay decreases by $\sim 56\%$. Hence, by assuming that $\beta = 0.56$, we estimate that the remineralization flux, $\beta C_{jss} V_{jss}$, equals $1.9 \times 10^{17}$ pmol yr$^{-1}$.

Estimating $F_{Nd}^j$ is more difficult, because information about $F_{Nd}^j$ is not immediately available in the literature. Only a few studies have reported Nd concentrations in Japan Sea benthic porewaters [e.g., 54], and estimates for $D_{Nd}$ require knowledge of the porosity and tortuosity of sediments as well as the distribution coefficient of Nd between porewater and sediment surfaces [e.g., 14,55,56]. As an alternative, we note that coastal sediments in the western North Pacific, which are a mixture of eolian and hemipelagic material from the Chinese Loess Plateau and circum-Pacific island areas, have an average Nd isotope composition ($\varepsilon_{Nd(sed)}$) of $\sim 4.2$ [20]. Thus, by assuming that $\varepsilon_{Nd(sed)} = -4.2$, it is observed that Eqs. (14) and (15) yield a system of two equations and two unknowns ($F_{Nd}^j$ and $k_{jss}^p$). In other words, the magnitude of $F_{Nd}^j$ must balance with the other inputs and outputs, including $k_{jss}^p C_{jss} V_{jss}$, so that the calculated value of $\varepsilon_{Nd(jss)}$ equals the measured value. This assumption yields a $F_{Nd}^j$ value $\sim 1.3 \times 10^{17}$ pmol yr$^{-1}$. We may check the quality of this estimate by normalizing $F_{Nd}^j$ to $A_{jss}$ and comparing the resulting flux to similar data reported for other settings. Our calculated value of $F_{Nd}^j$ is firmly within the range of $2.1–36$ pmol cm$^{-2}$ yr$^{-1}$ measured for the California and Chilean margins [14].

A maximum benthic input of $\sim 1.7 \times 10^{17}$ pmol yr$^{-1}$ is obtained by assuming that $\beta = 1.0$. Alternatively, with $\beta = 0.56$, Eq. (14) demonstrates that the theoretical minimum estimate for $F_{Nd}^j$ is $2.0 \times 10^{16}$ pmol yr$^{-1}$ (i.e., $k_{jss}^p = 0$). In this latter case, to match the predicted and observed $\varepsilon_{Nd(sed)}$ values, Eq. (15) requires that $\varepsilon_{Nd(ben)}$ equal $+1.0$, which is roughly the upper limit for the isotope composition of island arc rocks [5]. While this scenario may be plausible, the overarching implication from these calculations is that the benthic input of Nd is relatively minor compared to the input from calcite dust. It should be noted, however, that additional work is required to constrain $F_{Nd}^j$ and that the relative importance of atmospheric versus benthic inputs interpreted for the Japan Sea may not apply globally. In other
locations, where atmospheric inputs appear minimal, substantial benthic inputs are a key component of the seawater Nd budget [12,14–16]. By setting $\beta$ equal to 0.56 and $J_{\text{ben}}^{\text{Nd}}$ equal to 1.3 × 10$^{17}$ pmol yr$^{-1}$, we find that $k_{\text{jsd}}$ equals 1.9 × 10$^{-3}$ yr$^{-1}$. Thus, the particle scavenging flux of Nd ($J_{\text{jsd}}^{\text{Nd}}C_{\text{jsd}}V_{\text{jsd}}$) equals 1.1 × 10$^{17}$ pmol yr$^{-1}$, and the residence time with respect to particle scavenging ($\tau_{\text{jsd}}^{\text{Nd}}$) equals ~530 yr. This value is broadly comparable to the residence times of 200–500 yr reported for the global ocean [2,6,9]. Because the hydraulic retention time of water in the jsd($\tau_{\text{jsd}}^{H}$) equals ~130 yr, the jsd is likely very well-mixed with respect to Nd. According to Eq. (17), the total residence time of Nd in the jsd reservoir ($\tau_{\text{jsd}}^{\text{Nd}}$) is ~100 yr.

The model results presented above demonstrate that processes occurring in the surface layer of the Japan Sea are the primary factors for establishing the Nd composition of deep water, as opposed to processes occurring in deep water driving changes at the surface. We calculate that ~65% of the Nd in the jss originates from calcite dust, while ~26% originates from the jsd. In the jsd, ~77% of the Nd originates from the jss, which means that ~50% of the Nd in the jsd is from calcite dust. Because the benthic flux accounts for only ~23% of the remaining Nd in the jsd, we conclude that the benthic input of Nd exerts less control on the Nd chemistry of Japan Sea waters relative to the atmospheric input from calcite dust.

Lastly, recent studies have suggested that dissolution/exchange processes involving coastal sediment may be more important sources of Nd to the sea surface than atmospheric inputs [2,3,6,9]. However, in the case of the Japan Sea, the isotope mass balance presented in Eq. (13) does not support this assertion. As discussed previously, the Nd isotope composition of circum-Pacific coastal sediment ($\varepsilon_{\text{Nd(sed)}}$) is ~−4.2 [20]. If we assume that the entire Nd input of 1.1 × 10$^{18}$ pmol yr$^{-1}$ originates from coastal sediment as opposed to calcite dust, we observe from Eq. (13) that $\varepsilon_{\text{Nd(jss)}}$ equals ~−5.00. This estimate is far more radiogenic than the observed value of −7.83. Even if we use the minimum possible Nd input of ~7.6 × 10$^{17}$ pmol yr$^{-1}$ estimated from Eq. (12), the predicted value for $\varepsilon_{\text{Nd(jss)}}$ is only ~−5.20. In both cases, the discrepancy worsens if a more radiogenic value for coastal sediment is used, such as that expected for sediments shed directly from Japan. While these estimations do not definitively rule out a contribution from coastal sediment, it appears from Eqs. (12) and (13) that the more important source for the Japan Sea is calcite dust.

6. Conclusions

We showed that the atmospheric supply of calcite dust from the Chinese Loess Plateau to seawater exerts a critical control on the isotope composition and residence time of Nd in the Japan Sea basin. The wet deposition of dissolved calcite at the Japan Sea surface supplies 1.1 × 10$^{18}$ pmol yr$^{-1}$ of soluble Nd with an $\varepsilon_{\text{Nd}}$ value of ~−8.6. This input is required to produce the observed concentration and isotope composition of Nd in the basinal waters. The residence times of Nd in the Japan Sea surface and deep, with respect to particle scavenging, are ~14 and 530 yr, respectively. The combined effects of water mass turnover and particle scavenging yield Nd residence times in the surface and deep reservoirs of ~2.8 and 100 yr, respectively. A flux of Nd from benthic porewaters is required to balance the isotope composition of Nd delivered to the Japan Sea deep by downwelling surface water and particle remineralization. However, the benthic input is much smaller than the input from dissolved calcite dust. The isotope mass balance of the Japan Sea does not appear to support a significant input of Nd from coastal sediments, but this finding does not negate the importance of coastal sediment processes in other locations [2,3,10–12,17].

The data also reveal that ~65% of the Nd flowing into the North Pacific from the Japan Sea surface originates from the deposition of dissolved calcite dust. Mostly likely, the greater North Pacific also receives Nd directly from the input of dissolved calcite dust. However, it appears from the atmospheric transport model used in the study that the deposition rate of dissolved calcite east of the Japanese Islands is very low. The highest deposition rate occurs at the continental margin, which provides a reasonable explanation for why the isotopic signature of dissolved calcite dust is not immediately evident in the distal North Pacific [e.g., 20]. Neodymium in calcite dust contributes to the Nd inventory of the North Pacific, but only after it cycles through the Japan Sea. These observations support the contention that the cycling of Nd in marginal seas should be considered as an important link between terrestrial processes and open ocean chemistry [57].

Lastly, recent studies have reported that dust dissolution is capable of lowering the residence time of Nd in the global ocean to ~200–500 yr [6,8,9]. Our findings generally support this contention. However, we add the observation that dust cannot be treated as a uniform material. Ascribing a bulk dissolution rate to dust underestimates the complexities of the dust contribution. It is necessary to consider the abundances,
dissolution rates, transport mechanisms, and Nd geochemistry of the various mineral phases that comprise dust. Although calcite is a minor constituent of fresh Chinese loess (~10 wt.%), its rapid dissolution during atmospheric transport has a disproportionately large effect on the geochemistry of seawater, particularly near the Loess Plateau. The more abundant but generally less soluble phases in loess appear to provide a much smaller contribution. Similar cases may exist for other ocean basins proximal to arid regions.

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