Online supplement to

Holocene Evolution in Weathering and Erosion Patterns in the Pearl River Delta

1. Impact of carbonate strontium to the detrital $^{87}Sr/^{86}Sr$ ratios

We employed $^{87}Sr/^{86}Sr$ data measured from both bulk and decarbonated (silicate only fractions) sediment from ODP Site 1144 in the northern South China Sea and published by *Hu et al.* [2012] to estimate the impact of carbonate strontium on bulk sediment $^{87}Sr/^{86}Sr$ values in the Pearl River delta because these data allow us to estimate the impact of carbonate contamination on $^{87}Sr/^{86}Sr$ values and assess whether these can be used as a measure of chemical weathering intensity. The total inorganic carbon (TIC) data (Supplementary Fig. 1 A(2) and B(2)) measured by coulometry provides a good estimate of the carbonate content of the sediment and allow us to conclude that <1% (<0.4% in Pearl River delta) of CaO we measured by X-ray Fluorescence is not composed by detrital carbonates or biogenic carbonate and largely represents the presence of feldspar (Supplementary Fig. 1 A(3), B(3)). As described in the introduction section, carbonate in the Pearl River delta (not including the shells avoided for analysis) is eroded from Devonian limestone and minor Triassic dolomite that outcrop in the western part of the drainage system [*Chen et al.*, 2005].

The proportion of carbonate within the sediment is minor throughout the cored sections at HKUV-1 and B2/1, but is higher in the post 14 ka sediments analysed at ODP Site 1144 by *Hu et al.* [2012] because of the presence of carbonate microfossils (Taiwanese rivers which are the dominant supplier of clastic material do not provide detrital carbonate to ODP Site 1144).

Strontium from either biogenic or detrital carbonate sources yields lower values of $^{87}Sr/^{86}Sr$ compared to that measured from the weathered siliciclastic continental flux. As a result, the presence of carbonate reduces $^{87}Sr/^{86}Sr$ values when bulk sediment is measured without decarbonation. The $^{87}Sr/^{86}Sr$ values measured for bulk detrital sediment is a combination of the relatively higher values of the silicate components reduced by the presence of the carbonate component. To assess how much $^{87}Sr/^{86}Sr$ values are reduced by the presence of carbonate strontium, we modelled the bulk sediment to be only composed of silicate and carbonate components (Equation (1)). In this study, because of the relative short period of time for the initial differentiation of strontium isotopes in the continental carbonate rocks and biogenic calcite shells, these carbonate components are assumed to have remained constant in $^{87}Sr/^{86}Sr$ values through the study period. TIC data are used to calculate the content of carbonate, which, also, is assumed to have a steady concentration of strontium for all the sediments considered in this work. In turn the contribution that the strontium in the carbonate makes to the bulk $^{87}Sr/^{86}Sr$ values can be calculated by substituting (2) into (1).

$$^{87}Sr\,/\,^{86}Sr_{silicate} \times Sr\%_{silicate} + {}^{87}Sr\,/\,^{86}Sr_{carbonate} \times Sr\%_{carbonate} = {}^{87}Sr\,/\,^{86}Sr_{bulk} \times Sr\%_{bulk} \quad (1)$$

$$Sr\%_{carbonate} = TIC \times \frac{100}{12} \times Sr\%_{[in\ carbonate]} \quad (2)$$

Earlier studies have measured strontium concentrations of 1000 and 1700 ppm in detrital and biogenic carbonates respectively ($Sr\%_{[in\ carbonate]}$). These values are employed in this calculation [*Fantle and DePaolo*, 2006]. In the Pearl River delta, an average $^{87}Sr/^{86}Sr$ value of 0.7087 was determined from a study of the carbonate bedrocks in the western part of the river basin by *Chen et al.* [2005]. This value is used for determining the detrital carbonate fractions

($^{87}Sr/^{86}Sr_{carbonate}$). In the northern South China Sea, we use a biogenic carbonate ^{87}Sr/^{86}Sr value of 0.7092, taken from measurements in the western Pacific Ocean [*Hess and Schilling*, 1985].

We evaluate our model in the northern South China Sea by comparing the calculated silicate ^{87}Sr/^{86}Sr values to the measured results from ODP Site 1144 from the decarbonated fractions. Supplementary Figure 1A shows that calculated results (green stars) lie close to the measured values for the decarbonated fractions (red circles), and are distinct from the measured results from bulk sediment (blue circles) (Supplementary Fig. 1A(1)). This suggests that our model provides a reasonable estimate of the effect that carbonate has on bulk ^{87}Sr/^{86}Sr values. Supplementary Figure 1B shows the results of applying the same model to the Pearl River estuarine sediments, where carbonate is much less abundant. The calculated siliclastic ^{87}Sr/^{86}Sr values show variation with a departure of up to 0.04 greater than the measured results from the bulk sediment. The limited difference suggests that the TIC that reflects the presence of <4% carbonate in the Pearl River delta does not significantly change the overall trend of the siliclastic ^{87}Sr/^{86}Sr reconstruction (Supplementary Fig. 1B(1)). The presence of carbonate appears to have particularly increased ^{87}Sr/^{86}Sr values around 6 ka, suggesting that chemical weathering was weaker than would be interpreted from the bulk data at that time. Related TIC and CaO contents are plotted for reference (Supplementary Fig. 1 A, B (2–3)). Data are provided in Supplementary Table 2.

References

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Figure and Table Captions

Supplementary Figure 1. Part (A). Plots of northern South China Sea data show (1) comparison between calculated and measured $^{87}Sr/^{86}Sr$ values for siliclastic and bulk sediment; (2) TIC measured by columetry; (3) CaO measured by XRF and calculated from columetry TIC values. Part (B). Plots of Pearl River data show (1) comparison of $^{87}Sr/^{86}Sr$ values between calculated siliclastic and measured bulk sediment; (2) TIC measured by columetry; (3) CaO measured by XRF and calculated from columetry TIC values. In Part B circles represent data from Core HKUV1, squares from Core B2/1.

Supplementary Table 1. Results of major and trace element abundances in bulk sediment by XRF measurement at University of Massachusetts Boston, MA, USA. Major elements are given in oxides in weight percentages (wt. %) and trace elements in μg/g.

Supplementary Table 2. Data used for estimation of carbonate strontium isotope $^{87}Sr/^{86}Sr$ that contribute to the value of bulk sediment in both the north South China Sea and the Pearl River delta.