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Storage and release of fossil organic carbon related to weathering of sedimentary rocks

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Abstract

The biogeochemical carbon cycle, which plays an undeniable role in global climate change, is defined both by the size of carbon reservoirs (such as the atmosphere, biomass, soil and bedrock) and the exchange between them of various mineral and organic carbon forms. Among these carbon forms, fossil organic carbon (FOC) (i.e., the ancient organic matter stored in sedimentary rocks) is widely observed in modern environments but is not included in the supergene carbon budget. Using a digitized map of the world and an existing model of CO_2 consumption associated with rock weathering, we establish the global distribution of FOC stored in the first meter of sedimentary rocks and a first estimation of annual FOC delivery to the modern environment resulting from chemical weathering of these rocks. Results are given for the world's 40 major river basins and extended to the entire continental surface. With a mean value of 1100 10^9 t, mainly controlled by shale distribution, the global FOC stock is significant and comparable to that of soil organic carbon (1500 10^9 t). The annual chemical delivery of FOC, estimated at 43 10^6 t yr^{-1} and controlled by the areal distribution of shales and runoff, is of the same order of magnitude as the FOC output flux to oceans. Chemical weathering of bedrock within the Amazon basin produces one-quarter of the total global flux of FOC derived from chemical weathering, and thus is expected to govern FOC release on a global scale. These results raise important questions concerning the role of FOC in the modern carbon cycle as well as the origin and the budget of carbon in soils and rivers.

Keywords: global carbon cycle; spatial distribution; major river basins; weathering; fossil organic carbon flux; fossil organic carbon storage

1. Introduction

Fossil organic carbon (FOC) is derived from ancient organic matter (OM) (kerogen, in the language of petroleum geologists) buried with mineral matter in sedimentary basins. During burial, time, temperature, and pressure affect the chemical and physical properties of OM until a

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graphitic form is attained [1]. In the right geological setting, and especially where uplift has occurred, sedimentary rock bearing this ancient OM and its corresponding FOC can outcrop at the continental surface. FOC then can be delivered to modern environments through the mechanical erosion and chemical weathering of these sedimentary rocks (Fig. 1). Previously, the role of FOC in modern environments was considered only in the context of a carbon balance (i.e., to balance the burial of organic carbon through the total mineralization of FOC at outcrop), with an annual flux of 100 10^6 t, and hence to maintain a constant atmospheric level of O_2 and CO_2 [2]. However, because a part of this FOC escapes from diagenetic processes during the geological cycle, it is believed also to be partially recalcitrant to mineralization at outcrop.

FOC has been identified as being ubiquitous in rivers [3–6] and, more recently, in soils [7], and even in recent marine sediment [8,9]. In light of these observations, it is extraordinary that FOC has never been quantified in the supergene organic carbon budget (Fig. 1, [10–12]). Only two studies propose an annual estimate of FOC delivery to the world's oceans, one describing delivery by world river basins (80 10^6 t yr^{-1}, [13]) and the other by small mountainous rivers (40 10^6 t yr^{-1}, [14], Fig. 1). Other authors have suggested that on the order of 40–70 10^6 t yr^{-1} of FOC may be reburied [15]. There currently is no quantitative information available regarding the transport of FOC by rivers before its delivery to the oceans. Providing meaningful global-scale data concerning the contribution of FOC from the continental surface is not straightforward. This shortcoming is aggravated by the lack of distinction between FOC and black carbon (the combustion residue of fossil fuel and present vegetation [16]), which, to a certain extent, invalidates estimates of these types of inert carbon [8]. In determining the contribution of FOC to modern environments, the initial step in the delivery of FOC – i.e., its release through mechanical erosion and chemical weathering of sedimentary rocks from its storage in the upper part of these (sub)-outcropping rocks – must be considered (Fig. 1). One study claims that the chemical weathering of carbonates and shales produces 100 10^6 t yr^{-1} FOC [17], partitioned between soils, rivers, and, through mineralization, the atmosphere, and no data exist for the amount of FOC produced by mechanical erosion except for that produced by small mountainous watersheds [40 10^6 t yr^{-1}, 14].

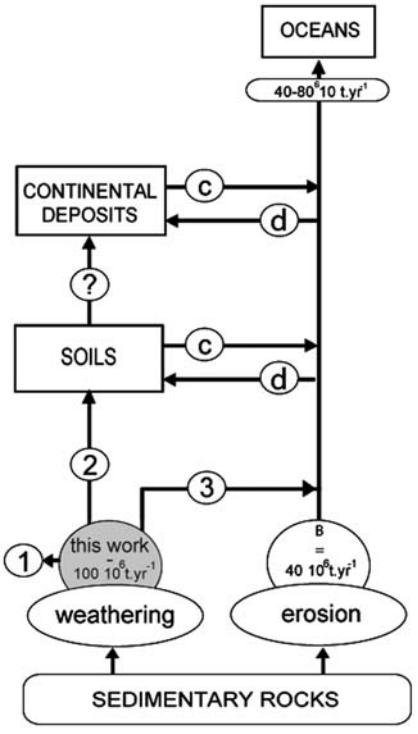

Fig. 1. Relations and exchanges of FOC within continental surfaces. The letters refer to terms in Eq. (6): FOC fluxes from weathering of rocks, from [17] and this study (*A*); FOC fluxes from erosion for mountainous coastal catchments [14] and other world rivers (not quantified) (*B*); FOC supplied from the remobilisation processes (*C*); deposition of FOC in continental reservoirs (*D*); mineralization occurring during transport of FOC (black arrows) and within reservoirs (*E*). FOC main fate processes after release by chemical weathering indicated by (1) mineralization; (2) input to soils; (3) direct input to rivers.

Here we present an estimate of the FOC flux contributed by the chemical weathering of sedimentary rocks. We use a GIS survey to estimate the original FOC stock contained in the upper layers of sedimentary rocks for the world's 40 major river basins, and extend it to the entire continental surface. We then estimate the FOC flux attributable to the chemical weathering of carbonates and shales, and its distribution for the watersheds studied and on a global scale. These initial results might provide a useful starting point for further investigations concerning the contribution of FOC to continental surfaces and hence to the supergene carbon cycle.

2. Methods

The general approach used here was to calculate the amount of FOC in storage and the FOC yield originating from the chemical weathering of shales and carbonates for the whole continental surface, and then to calculate average budgets for the world's 40 major river basins. Numerical maps of FOC storage and FOC chemical yield were established at the global scale using GIS software at a $1° \times 1°$ grid resolution and world maps of river basin limits, continental rock lithology [18], and continental runoff [19]. The global distribution of the lithology used