Yale 耶鲁大学-南京信息工程大学大气环境中心
Yale-NUIST Center on Atmospheric Environment

A discussion on the paper "The composition and transport of organic carbon in rainfall : Insights from the natural (^{13}C and ^{14}C) isotopes of carbon"

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About

The Raymond lab's research focuses on biogeochemistry of natural systems. In particular, we are interested in the carbon and nitrogen cycles within aquatic systems. Current research topics include the landscape controls on the watershed export of carbon, biogeochemical transformations in estuaries, the physics of air-sea CO2 exchange, nitrogen cycling in temperate watersheds, and determining the age and composition of carbon being transported from land to the ocean. Our research often utilizes the watershed approach and natural isotopes to determine major sources, sinks, and ages of various carbon and nitrogen pools in the natural environment.

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Introduction

- The sources and composition of OC in rainwater are unknown, leading to its uncertainty role in regional and global C budgets.

- **What we know is that:**

✓ The net flux of water <0, but the carbon flux in precipitation is in the opposite direction.

✓ There are appreciable transfers of marine OC between marine ecosystems and to the continents.

✓ Organic compounds originated from fossil fuel burning was found in the atmosphere and in rainwater.

Introduction

- **What is lacking:**

✓ Detailed knowledge about the transfers of OC through precipitation.

✓ Studies on the quantities, sources and reactivity of organic matter in rainfall.

- **The study was hampered by:**

✓ The low concentrations and chemical complexity of OC in rainwater.

Materials and methods

- **Aim of the experiment:**

  To assess the quantities and sources of rainwater OC.
- **Two sites**: New Haven, CT a small city on the Long Island Sound & Woods Hole MA,NE of US

- **Experiment procedures:**

  Collection→Transfer→Frozen(not filtered) → Transfer to a quartz reaction tube → Acidified → Sparged with UHP N_2 → Oxidize using a high energy UV lamp → Resulting CO_2 was then cryogenically purified → Isotopic analysis

Results and discussion

- Multiple sources of OC including a $\delta^{13}C$ enriched and a $\Delta^{14}C$ depleted relic contribution are apparent.
- Previous studies reported two :<u>compounds from contemporary vegetation emissions</u> and <u>relic OC from fossil fuel burning.</u>
- For $\delta^{13}C$ enriched samples：

✓ Marine biota produce significant quantities of VOC that contribute to atmospheric OC pools and marine aerosols associated with seaspray.

✓ Studies have demonstrated that rainwater originating over the ocean and sampled before contact with land contains a small amount of OC seemingly from marine sources.

Table 1. Rainfall Amounts, Concentrations, Isotopic Ratios, and Relative Contributions of the Three Major OC Sources to Rainfall Events Estimated Using a Dual Isotopic Mixing Model[a]

Date	Rain, In	OC, μM	$\delta^{13}C$, ‰	$\Delta^{14}C$, ‰	^{14}C-age	% Marine	% Fossil Fuel	% Modern Continent.
03/28/01[b]		58	−25.0	−441	4657	45	45	10
09/23/03	0.9	28	−24.9	−188	1727	35	23	42
10/27/03	1.4	119	−28.5	−653	8567	0	66	44
10/29/03	1.2	42	−25.9	−290	2806	21	34	45
11/05/03	0.3	100	−25.9	−95	850	12	17	71
12/11/03	0.5	34	−26.8	−191	1752	1	27	72
02/06/04	1.7	33	−25.8	−14	169	10	10	80
04/01/04	0.7	69	−24.8	108	Modern	21	0	79
04/12/04	3.8	30	−20.1	−160	1450	96	4	0
09/17/04	4.1	28	−22.5	−150	1362	76	16	8
09/28/04	2.8	30	−21.6	93	Modern	78	0	22
AVERAGE		52	−24.8	−180	2122	36	22	43

[a]See auxiliary material.
[b]Collected from Woods Hole, MA; all other samples from New Haven CT.

- 3 dominant sources of organic carbon to the rain samples collected by this study are modern terrestrial vegetation, fossil fuels, and marine OC sources.

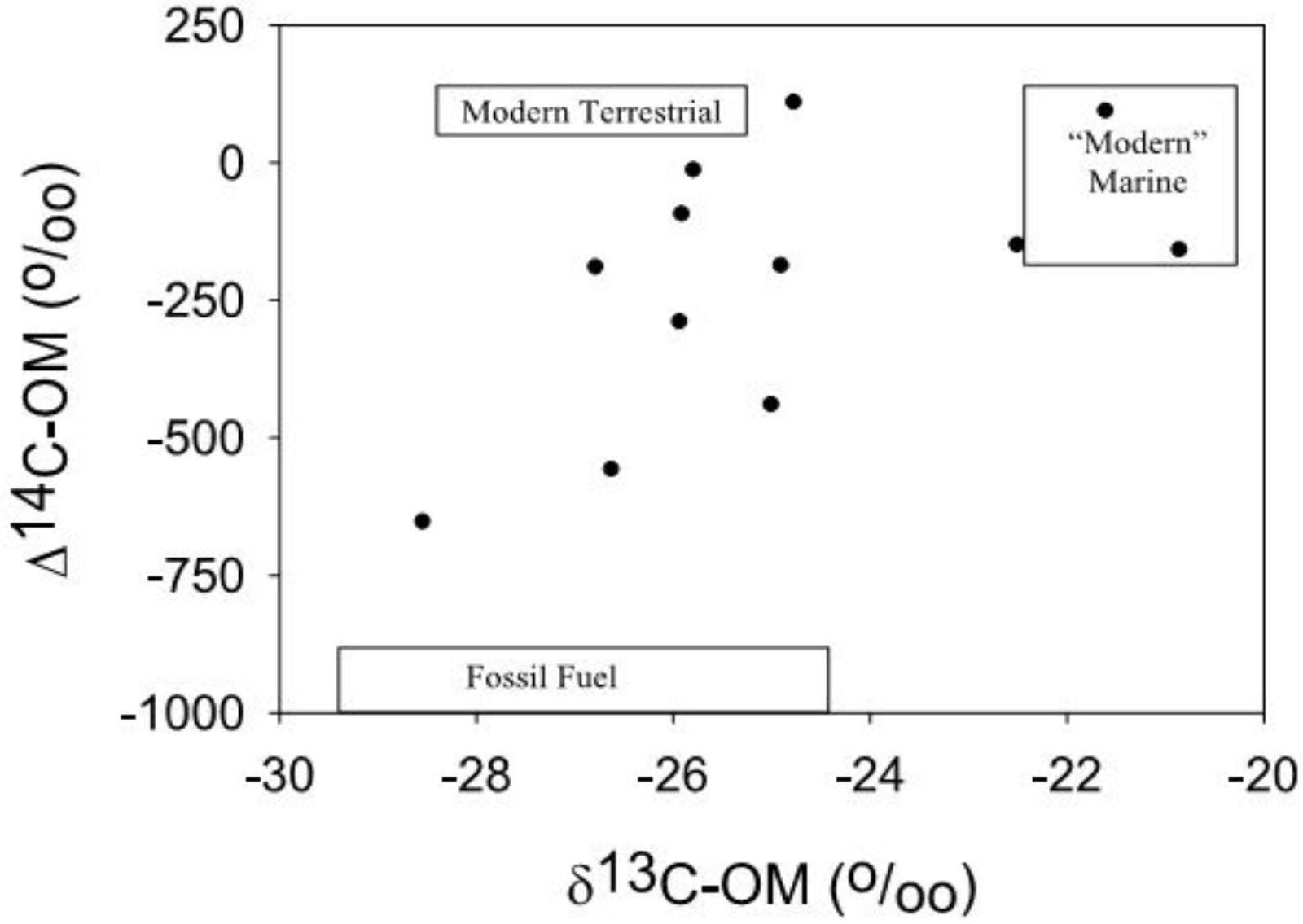


Figure 1. An isotopic source diagram for all rain samples.

Results and discussion

- For $\Delta^{14}C$ depleted relic samples：
- Small global pool size of fossil fuel OC

  ⇒ its transfer through wet deposition could be a considerable component of the relic carbon budget.
- Large ^{14}C-depleted OC reservoirs was found and consist of a large modern pool and a small ^{14}C-depleted relic pool .
- Determining the controls on the input, removals and transfers of relic carbon budget is important .

- The three end-member calculation estimates that ~20–30% of rainwater OC is relic carbon.
- Highly populated urbanized areas represent a direct source of relic OC to the coastal waters and the phenomenon is not limited to the coastal Atlantic.
- Marine OC transfer to land:

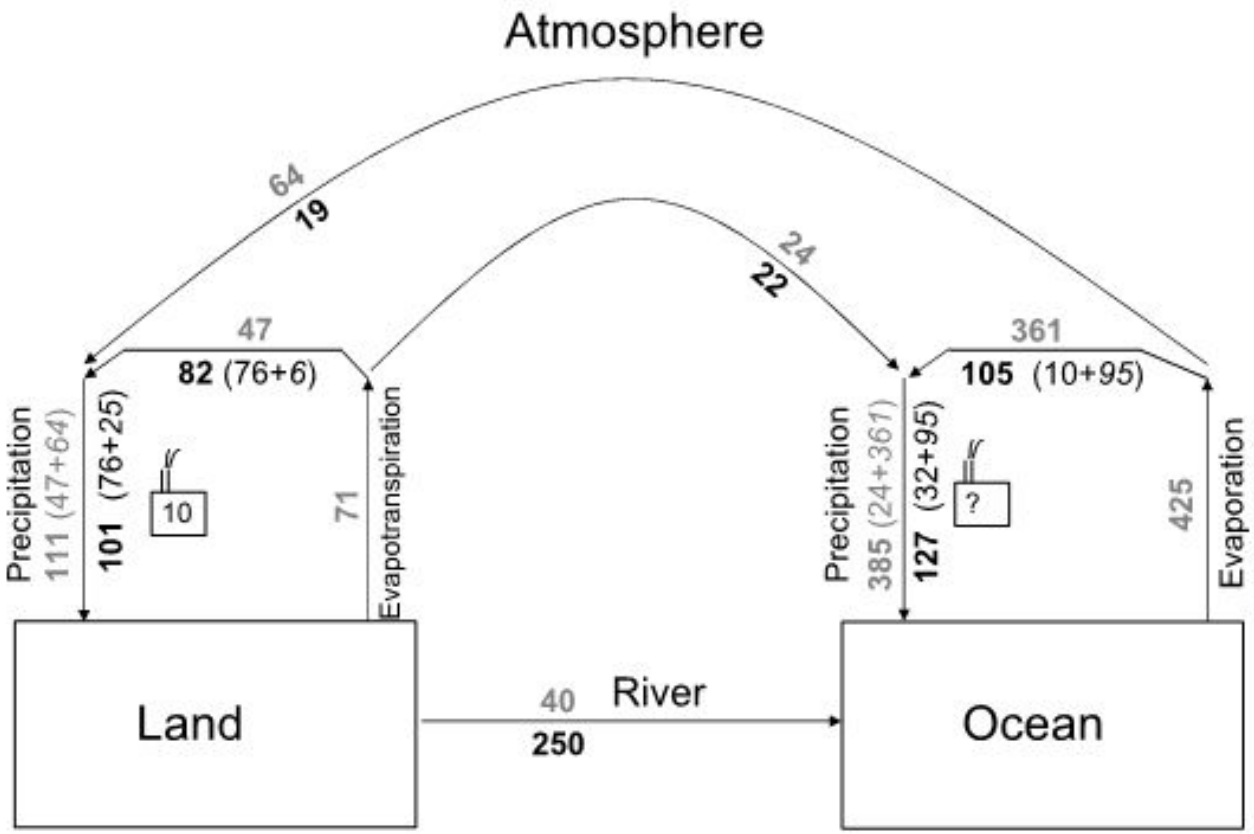
✓ All three marine OC dominated stoms traveled >2000km over land , they all had intact marine OC signatures and presumably deposited marine OC over the entire track.

✓ Coastal sites showed low concentrations of OC during hurricanes and ocean originating storms and this material can be almost entirely marine OC

⇨ *Storms have the ability to move marine OC far inland.*

- Regional and global carbon budgets have recently begun to assess the potential importance of the transport of terrestrial organic carbon in rainwater as a source of reduced carbon to the oceans.
- OC in storms originating over the ocean can be dominated by marine OC ⇨ marine OC recycling ⇨ transferring marine OC between marine ecosystems
- Future research should determine if specific marine ecosystems receive a subsidy of wet deposition OC from neighboring or even distant marine systems.

The major assumption:the gross water transfers are accompanied by OC fluxes in proportion to the concentration and source apportionments obtained by this study.



These estimates are first order and the transfers of OC may be decoupled from the transfers of water.

Figure 2.A conceptual diagram for the hydrologic transfers of water and organic carbon between the continents and ocean.

- It is difficult to accurately assess how much OC is exchanged between the terrestrial and oceanic basins via precipitation
- **The auther's point of view:**

✓ A large component of OC in marine rainwater is probably of marine origins.

✓ Net flux of OC from the continents to the ocean is smaller due to an appreciable ocean to land flux.

✓ Oceanic studies that invoke an allochthonous input of terrestrial OC via rainfall must also consider an output.

Conclusions

- OC in rain originates from multiple sources including a significant fraction from fossil fuels and marine OC.
- Atmospheric wet deposition of OC from fossil fuels can supply significant quantities of relic carbon to ecosystems.
- There is a significant transfer of marine OC to the continents and a large recycled marine OC component to marine rain.
- The net transfer of continental OC to the ocean could be close to zero due to a significant marine to continental flux.

Current work

- 雨水中颗粒物的研究：
- 主要方向：成分分析、溯源、雨水对污染物的清除作用（结合大气颗粒物的研究）
- Where we are:

  最初的试验设计、购买仪器等
- 雨水采集（APS-3A）→分装（酸洗过的PE瓶）→测定样品PH、电导率值→过滤（溶剂过滤器）→后续成分分析

Thanks for the time!