



Upper-ocean gas dynamics from radon profiles in the Eastern Tropical South Pacific



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ABSTRACT

Uncertainties in the dynamics of dissolved gases limit the accuracy of geochemical primary-productivity estimates. Mixed-layer ventilation, entrainment, and upwelling all affect the interpretation of geochemical data, yet they are rarely measured together. We report upper-ocean dissolved-gas dynamics in the Eastern Tropical South Pacific (10–20°S, 80–100°W) obtained from ²²²Rn distributions from a research cruise in February 2010. Radon-222-based surface ventilation measurements at seven stations were compared with gas transfer velocities calculated using empirical wind-speed parameterizations. Good agreement between the two methods was observed across different wind data products (ASCAT scatterometer and NCEP/NCAR meteorological reanalysis) and grid scales (0.5° × 0.5°, 1° × 1°, and 2° × 2°). Average wind speeds in the region were 5–8 m s⁻¹, corresponding to in situ gas transfer velocities of 1–3 m d⁻¹. Averaged over the region, most recent gas exchange parameterizations performed similarly, and the results are robust with respect to both wind data products and grid scales. Non-steady-state conditions were observed at one station, possibly caused by a recent internal wave. These results suggest that ²²²Rn could be used to support measurements of biogeochemical O₂ and CO₂ fluxes in the upper ocean.

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1. Introduction

The distribution of biologically relevant gases (e.g., O₂, N₂, and CO₂) and their isotopic composition in the mixed layer is the basis for a powerful class of approaches for studying sea-surface biogeochemistry, and in particular, primary productivity (Bender and Grande, 1987; Craig and Hayward, 1987; Emerson, 1987; Hamme et al., 2012; Hendricks et al., 2004, 2005; Juraneck and Quay, 2005, 2010, 2013; Luz and Barkan, 2000; Munro et al., 2013; Nicholson et al., 2010; Quay et al., 1993, 2010, 2012; Reuer et al., 2007; Stanley et al., 2010; Yeung et al., 2012). Recent methodological and technological advances (Cassar et al., 2009; Sarma et al., 2005) have made possible in situ studies of primary productivity with high spatial resolution and/or large areal extent. Although free of the potential artifacts that may bias on-deck incubation experiments, in situ methods still have well known uncertainties. Principal among them is the air-sea gas transfer velocity, k (m d⁻¹), which is important for understanding the balance between biological activity and the physical transport mechanisms that govern the mixed-layer budget of dissolved gases.

Wind speed is widely recognized as a robust parameter from which the gas transfer velocity can be calculated (Asher and Wanninkhof, 1998; Wanninkhof et al., 2009). It is convenient for field studies because accurate wind-field data are readily available, e.g., from satellite-based scatterometer instruments or meteorological data; therefore, wind-speed parameterizations require no additional experimental effort on research cruises. In contrast, other methods to estimate transfer velocity such as tracer injection (Ho et al., 2006; Nightingale et al., 2000; Watson et al., 1991), direct-covariance flux (McGillis et al., 2001, 2004), and passive gas flux methods (Sarma et al., 2010; Stanley et al., 2009) generally require a significant experimental effort, optimal field conditions, or both, to obtain accurate gas-transfer velocities. The trade-off for methodological simplicity, however, is uncertainty: Because wind-speed parameterizations may not explicitly account for complicating factors that vary in time and space, e.g., bubble injection, uncertainties in the best parameterizations are currently estimated to be about ±20% (2σ) over 1–3-day timescales (Ho et al., 2006, 2011). Moreover, surface ventilation is not the only factor affecting the dissolved-gas budget in the open ocean.

Other physical factors affecting the dissolved gas budget of upper-ocean water masses include short-term perturbations such as horizontal advection, internal waves, entrainment, and rapid changes in the mixed-layer depth. These influences violate typical

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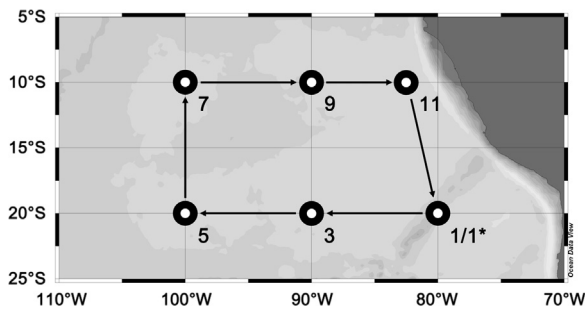


Fig. 1. Map of study area and steaming directions in the Eastern Tropical South Pacific. Station numbers are noted.

assumptions of a steady state between surface ventilation and diffusion-driven water-column transport (Kaiser, 2011; Nicholson et al., 2014, 2012). They are difficult to identify a priori, especially without separate tracers of water-mass history. Non-steady-state effects may be mitigated somewhat when regional estimates of biogeochemical parameters are sought, but direct observations of their effects on the upper-ocean dissolved-gas budget would be useful.

The ^{222}Rn -deficit method may be able to diagnose instances when non-steady-state factors are important to the upper-ocean dissolved-gas budget. It uses a naturally-occurring radiogenic isotope, ^{222}Rn (mean lifetime $\tau=5.5$ days), to trace dissolved-gas dynamics over timescales of days to weeks. Its classical application was to air-sea gas exchange (Peng et al., 1979, 1974; Smethie et al., 1985), but the approach had notable shortcomings, namely, its sensitivity to horizontal advection and water-mass entrainment/detrainment (Roether and Kromer, 1984). Here, we instead exploit this sensitivity of the ^{222}Rn inventory to changes in mixed-layer dynamics and composition. We hypothesize that it can identify non-steady-state influences directly relevant to dissolved-gas budgets of biogeochemical importance in the mixed layer. We applied this approach to the upper ocean of the Eastern tropical South Pacific (ETSP; see Fig. 1).

We use the classical ^{222}Rn -deficit method to determine gas transfer velocities under the steady-state assumption at six stations in the ETSP during February 2010, and we compare the results with those derived from wind-speed parameterizations. We examined the sensitivity to the choice of parameterization, as well as to grid size and wind-field product. Within this framework, consistency between top-down (wind-speed) and bottom-up (^{222}Rn -deficit) approaches to surface ocean ventilation indicates that the surface dissolved-gas budget was approximately in steady-state during the integration time of the ^{222}Rn -deficit method. At one such station, we show that the ^{222}Rn inventory can constrain eddy diffusivities in the upper thermocline to within an order of magnitude.

The ETSP is defined here as the region between the zone of equatorial upwelling influence and the South Pacific Gyre—roughly between 5°S and 20°S (Pennington et al., 2006). It is a region where N_2 fixation and denitrification may be spatially coupled, thus playing an important role in regulating the global oceanic N budget over time (Deutsch et al., 2007; Westberry and Siegel, 2006). Furthermore, it is linked to the world's largest low-oxygen area, which lies in the upper thermocline at depths of only a few hundred meters (Codispoti et al., 1986; Fiadero and Strickland, 1968). The ETSP has remained relatively undersampled by field studies, however, in part because the potential importance of its biogeochemistry to the global nutrient budgets of the ocean has only recently been recognized. This study examines an important and understudied region of the ocean, and it constitutes a test case for using the ^{222}Rn -deficit to support studies of mixed-layer biogeochemistry.

2. Theory and methods

2.1. Boundary-layer diffusion paradigm for gas exchange across the air–water interface

The boundary-layer diffusion model is a basic physical model for gas exchange across the air–water interface, and it is the conceptual basis for both the ^{222}Rn -deficit and wind-speed methods. It is based on the assumption that molecular transport dominates mass transport at an interface, while turbulent transport dominates transport away from an interface (Jähne and Haussbecker, 1998; Prandtl, 1925). In the liquid phase, molecular diffusion is the rate-limiting process for transport, while in the gas phase, molecular diffusion and viscous (momentum) diffusion are comparable. This difference in mass transport properties results in water-side control for exchange of sparingly soluble gases (e.g., Rn , O_2 , N_2 , and Ar) and air-side control for exchange of highly soluble gases (e.g., SO_2). Next to solubility, wind is the most important environmental forcing on air-sea gas exchange because of its direct effects on the aqueous diffusive boundary layer.

2.2. In situ gas transfer velocity with the ^{222}Rn -deficit method

The ^{222}Rn -deficit method employed in this study utilizes the radioactive decay of ^{222}Rn as a 'clock' to trace physical influences on its inventory (Bender et al., 2011; Broecker et al., 1967; Broecker and Peng, 1971; Emerson et al., 1991; Peng et al., 1979, 1974; Roether and Kromer, 1978, 1984; Smethie et al., 1985). Produced by the α -decay of the well-mixed nuclide ^{226}Ra ($\tau\sim 2300$ years) in the water column, ^{222}Rn in the ocean interior reaches a steady-state concentration at equilibrium such that its activity is equal to that of ^{226}Ra . In the upper ocean, ventilation to a nearly radon-free atmosphere contributes a second loss term to the ^{222}Rn activity budget. If the system is at steady state, the magnitude of disequilibrium is directly proportional to the steady-state gas transfer velocity. In practice, radon depth profiles represent the integrated gas transfer velocity over 2–3 decay lifetimes (~ 2 weeks) if other physical factors are not significant (Bender et al., 2011; Roether and Kromer, 1984; Smethie et al., 1985). This gas transfer velocity k_{Rn} can be expressed as

$$k_{\text{Rn}} = \left(\frac{A_E}{A_M} - 1 \right) \lambda h \quad (1)$$

when the atmospheric contribution to the upper-layer ^{222}Rn activity is negligible. Here, A_E is the ^{222}Rn activity at equilibrium (in the absence of gas exchange), A_M is the ^{222}Rn activity at the surface, λ is the first-order decay constant of ^{222}Rn (0.1813 d^{-1}), and h is the equivalent depth of the ^{222}Rn deficit, i.e.,

$$h = \frac{\int_0^\infty (A_E - A_z) dz}{A_E - A_S} \quad (2)$$

where A_S and A_z are the ^{222}Rn activities measured at the surface and at depth z , respectively. This method was used extensively for gas exchange estimates during the GEOSECS program (Peng et al., 1979), as well as for the JASIN, FGGE (Roether and Kromer, 1984), and BOMEX programs (Broecker and Peng, 1971), and as part of a long-term observational program at Station PAPA (Emerson et al., 1991; Peng et al., 1974). It has well known limitations that have been discussed at length previously (Roether and Kromer, 1984), but a recent reanalysis of the GEOSECS dataset suggests that the method may not be as inaccurate as previously thought (Bender et al., 2011).

2.3. ^{222}Rn and ^{226}Ra analyses

Radon-222 depth profiles were measured onboard the *R/V Atlantis* from Niskin casts at seven stations encompassing the oceanic region bounded by 10–20°S and 80–100°W during a research cruise in February 2010 (see Fig. 1). This region includes the transition zone

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