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COURSE ON CLIMATE VARIABILITY
STUDIES IN THE OCEAN
"Tracing & Modelling the Ocean Variability"
16 - 27 June 2003

301/1507-7

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^{137}Cs and ^{239}Pu as new numerical tracers in an ocean general circulation model

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Abstract

We have carried out first simulation of the spatial distributions and the temporal variations of ^{137}Cs concentrations in the North Pacific in off line calculations by using archived output of an ocean general circulation model (OGCM) developed by the National Center of Atmospheric Research (NCAR). Artificial radionuclides including ^{137}Cs are introduced into ocean surface due to global fallout originating from the large-scale atmospheric nuclear weapons tests in 1961-62. The distribution of radioactive deposition used as forcing for this simulation is estimated from global precipitation data and observed values of annual deposition of radionuclides at the Meteorological Research Institute (MRI) in Japan. ^{137}Cs originating from global fallout have been transported into the ocean interior by advection and diffusion, and the ^{137}Cs concentrations reduced by radioactive decay. We assess the skill of the model calculations by comparing simulated values of ^{137}Cs in seawater with the observed values included in the database compiled by MRI because ^{137}Cs is one of the most useful tracers regarding water motion in the ocean. The vertical and horizontal distributions of the calculated ^{137}Cs concentrations were in good agreement with those of the observed ^{137}Cs concentrations, except in the deep layer.

In addition, we have also performed simulation of ^{239,240}Pu with considering simple scavenging effect. A maximum ^{239,240}Pu concentration layer occurs at intermediate depth for both observed and calculated values, which is formed by particle scavenging. The horizontal distributions of the calculated ^{239,240}Pu concentrations in surface water could be simulated by considering the scavenging effect.

In future, we plan online calculation for ^{137}Cs and ^{239,240}Pu by considering inter-annual variability. At fitst, using a ealistically forced, global 3-D numerical ocean general circulation model (OGCM), we examine how well CFC tracer age approximates "ideal age" in the North Pacific thermocline as a function of time and explore the magnitude of interannual variability in both age measures.

Contents

1. Introduction

The presence of artificial radionuclides in the North Pacific surface water has been derived largely from global fallout from atmospheric nuclear weapons tests together with a nuclear reactor accident at Chernobyl. The distributions of these radionuclides have been investigated to understand the transport of radionuclides in the ocean (Bowen *et al.*, 1980; Nagaya and Nakamura, 1981; 1984; 1987; Aoyama and Hirose, 1995; Livingston *et al.*, 2001). The Meteorological Research Institute (MRI) in Japan has collected historical data (Historical Artificial Radionuclides in the Pacific Ocean and its Marginal Seas (HAM) database, version U, Aoyama and Hirose, submitted(a)) on three anthropogenic radionuclides (^{90}Sr, ^{137}Cs and ^{239,240}Pu(The sum of ^{239}Pu and ^{240}Pu activities was described as ^{239,240}Pu in the database)) in the world ocean, mainly in the Pacific Ocean and its marginal seas. This database describes the spatial and temporal characteristics of global distribution in the ocean (Aoyama and Hirose, submitted(b)) and geographical distribution of decadal changes of surface radionuclides concentrations in the Pacific Ocean.

The depositions of artificial radionuclides originating from the atmospheric nuclear weapons tests, have been measured at many land-base stations around the world. In Japan, monthly measurements of ^{90}Sr, ^{137}Cs and ^{239,240}Pu depositions have been performed since 1957 on site at the MRI(Katsuragi *et al.*, 1982; Katsuragi, 1983; Hirose *et al.*, 1987; Igarashi *et al.*, 1996). However, there has been no measurement of the deposition of artificial radionuclides onto the open ocean, except for island stations. Sarmiento and Gwinn (1986) estimated the spatial distribution of ^{90}Sr depositions in the Atlantic Ocean using air concentrations and precipitation data. In this study, we estimated the spatial distribution of ^{137}Cs depositions onto the ocean considering the precipitation distribution over the Pacific Ocean and the meridional change of the concentrations of the radionuclides in rain water evaluated from land based observations.

Observations of chemical tracers, such as CFCs, tritium and ^{14}C, are useful to understand the behavior of materials in the ocean and to provide a measure of the accuracy of ocean general circulation models(OGCMs) (England and Maier-Reimer, 2001). Observed data is generally too sparse to fully describe the distributions of artificial radionuclides in the ocean. It is possible to interpolate and extrapolate observed data by an OGCM for the understanding of the behavior of artificial radionuclides in the ocean. In this paper, we show that ^{137}Cs is also a useful tracer for assessing the accuracy of OGCMs.

In addition, the acquired three-dimensional distributions of ^{137}Cs by this simulation provide insight into the fate of fallout in the ocean for assessment of the environmental impacts. Calculated spatial distributions of artificial radionuclides and their temporal change can be compared with observed data in the HAM database.

OGCMs have been developed by many institutes to study the transport of materials in the ocean and to predict future climate changes. Recently, OGCMs have greatly advanced as a result of the progress of physical oceanography and high performance computers. For example, the distribution of CFCs simulated by many OGCMs, were compared with observations and other models in the Ocean Carbon Cycle Model Inter-Comparison Project 2(OCMIP-2). In the OCMIP-2, input conditions are the same for all models, but there are significant differences of simulated results, which are caused by the differences of the flow field and the diffusion modeling between the models (Dutay *et al.*, 2002).

^{137}Cs is recognized as another useful tracer, which has a different input condition than CFCs. The numerical simulation of ^{137}Cs concentrations has been carried out to test a circulation model of the Black Sea (Staneva *et al.*, 1999). However, ^{137}Cs has not been used to assess OGCMs for the open ocean because of a lack of appropriate dataset of observed values. The compilation of the database for these isotopes by MRI (Aoyama and Hirose, submitted (a)) makes it possible to begin using them for testing ocean models.

In this paper, we present the first simulation of the spatial distributions and the temporal variations of ^{137}Cs concentrations in the ocean from offline calculations using archived flow fields of the NCAR Climate System Model (CSM) Ocean Model (NCOM)(NCAR Oceanography Section, 1996; Gent *et al.*, 1998). Figure 1 shows the schematic drawing of our model. Artificial radionuclides introduced into the ocean surface by global fallout with considering horizontal distribution are mixed homogeneously over the depth of the surface mixed layer. ^{137}Cs originating from global fallout are transported into the ocean interior by advection and diffusion, and these concentrations reduce by radioactive decay of 30.0 years half life. We assessed the results of the model calculations by comparison with the observed values included in the HAM database. In this study, the distributions of artificial radionuclides were analyzed only in the North Pacific because there are a large number of observations in the North Pacific in the HAM database and greater uncertainty in the input condition in the Southern Hemisphere.

2. Methods

2.1 Input to ocean

The deposition of artificial radionuclides on the ocean surface is extrapolated from the observed deposition data according to:

$$F(\lambda,\phi,t) = F_0(t)\frac{P(\lambda,\phi)}{P_0(t)}\varepsilon(\phi) \qquad \text{-----(1)}$$

where, $F(\lambda,\phi,t)$ is the estimated annual deposition(Bq m^{-2} yr^{-1}) on the ocean surface at the point(λ , ϕ), λ and ϕ are the longitude and latitude, respectively, t is time, $F_0(t)$ is the observed annual deposition(Bq m^{-2} yr^{-1}) on a reference site of MRI, $P(\lambda,\phi)$ is the annual precipitation(mm yr^{-1}) on the ocean surface at the point(λ , ϕ), $P_0(t)$ is the observed precipitation(mm yr^{-1}) on a reference site, and $\varepsilon(\phi)$ is the empirical function of the meridional distribution of concentrations of radionuclides in rain water.

The resulting meridional distribution of ^{137}Cs deposition is compared with previous land based estimates although geographic distribution of ^{137}Cs deposition on the ocean has still been unknown.

2.2 Results of Ocean General Circulation Model (OGCM)

We employed achieved results of the NCOM (NCAR Oceanography Section, 1996; Gent *et al.*, 1998) in an offline calculation of the concentrations of artificial radionuclides in the ocean. NCOM is based on Modular Ocean Model (MOM) version 1.1 (Pacanowski *et al.*, 1993).

2.3 Calculation of radionuclides concentrations in the ocean

The spatial and temporal evaluation of the tracers is governed by the advection - diffusion equation including radioactive decay:

$$\frac{\partial C}{\partial t} + L(C) = R(A_I, A_D, C) - \lambda_r C \quad , \qquad (2)$$

where C is the artificial radionuclide concentration (Bq m^{-3}), t is time (s), A_I and A_D are the isopycnal and diapycnal diffusivity ($m^2 s^{-1}$), respectively, λ_r is the decay constant of the radionuclides (s^{-1}), 2.31 x 10^{-2} (half-life time; 30 years) for ^{137}Cs.

The advection term $L(C)$ is

$$L(C) = \frac{1}{a\cos\phi}\left[\frac{\partial}{\partial\lambda}(uC) + \frac{\partial}{\partial\phi}(v\cos\phi C)\right] + \frac{\partial}{\partial z}(wC) \qquad (3),$$

where a is the radius of the earth, u, v *and* w are the zonal, meridional and vertical advective Eulerian velocities (m s^{-1}) based on the results of NCOM, respectively. Equation 2 was solved by the finite difference methods with time step, 0.0073 days.

3. Results and discussion

3.1 Horizontal distributions of ^{137}Cs depositions

Annual mean ^{137}Cs deposition from 1958 to 2000 was specified as input conditions.

3.2 Concentrations in the ocean

3.2.1 Horizontal distribution of ^{137}Cs concentrations

The observed ^{137}Cs concentrations in surface waters of the western North Pacific are lower than eastern ones in the 1960s, although depositions of ^{137}Cs to the western North Pacific surface waters were larger than that in the eastern one. Calculation of Exp-Ann dose not reproduce the observed distribution of the surface ^{137}Cs concentrations in the North Pacific in the 1960s. In contrast, the zonal distribution of the annual mean ^{137}Cs concentration calculated by Exp-Mon in 1963 coincides with that of the observed surface ^{137}Cs concentrations, which showed low in west and high in east of the North Pacific. The surface ^{137}Cs concentrations in the western North Pacific mid-latitude region decrease as a result of large scale mixing in winter. The formation of the deep mixed layer in winter in the western North Pacific mid-latitude region and shallow mixed layer throughout the year in the eastern one is thus an important process for reproductions of the observed surface ^{137}Cs distribution.

The surface ^{137}Cs concentrations in the 1970s decreased all sites in the North Pacific comparing with that in the 1960s. The highest observed ^{137}Cs concentrations occurred in the wide area off the western coast at the United States in the 1970s. This patch with relative high surface ^{137}Cs was observed by Saruhashi *et al.* (1975). Vdovenko *et al.* (1970) suggested that this patch was probably formed due to the effluent of radioactive wastes through the Columbia River. On the other hand, Folsom (1980) suggested an alternative hypothesis that a large amount of deposited ^{137}Cs in the western North Pacific was advected to the eastern North Pacific on a timescale of a decade. The geographic ^{137}Cs distributions are not qualitatively affected by the monthly change of mixed layer depth in the 1970s, because ^{137}Cs depositions in the 1970s were more than one order of magnitude lower than in the 1960s (Hirose et al., 1987). This model can realize the high ^{137}Cs concentrations patch only from the fallout input without the input from the Columbia River, supporting the hypothesis of Folsam (1980).

The surface ^{137}Cs concentrations in the 1980s are geographically homogeneous comparing with that in the 1960s and 1970s.

3.2.2 Vertical distributions of ^{137}Cs concentrations

The vertical distributions of the observed ^{137}Cs concentrations in each decade from the 1960s to the 1990s in the area (20 degree N ~ 40 degree N, 130 degree E ~ 160 degree E, excluding the Japan Sea) near Japan are compared with the calculated ones. The calculated vertical ^{137}Cs distributions in the area are in good agreement with the observed profiles in the 1960s up to 250m, in the 1970s up to 500m, in the 1980s up to 750m and in the 1990s up to 750m. The calculated results in the 1960s indicated that vertical ^{137}Cs profiles by Exp-Ann are different from one by Exp-Mon. Exp-Mon including the monthly change of mixed layer depth better reproduces the ^{137}Cs concentrations in surface and subsurface waters comparing with observed data for especially shallower than the depths of 250m in contrast of Exp-Ann. The better agreement of Exp-Mon is due to the monthly changes of mixed layer depth during high deposition period in this area in the 1960s.

4. Application for $^{239,240}Pu$ calculation

4.1 Simple scavenging model

The basic equation on the three-dimensional diffusion equation was taken into consideration both nuclides decay and scavenging, which is the removal of radionuclides in seawater by absorption onto suspended materials that deposit on the ocean floor by sedimentation. The basic equation is as follows;

$$\frac{\partial C}{\partial t} + L(C) = R(A_I, A_D, C) - \lambda_n C - K_d \rho_s(z) w_s \frac{\partial C}{\partial z} \quad , \qquad (2)$$

where C is the radionuclide concentration (Bq m^{-3}), t is time (s), A_I and A_D are the isopycnal and diapycnal diffusivity ($m^2 s^{-1}$) respectively, λ_n is the decay constant of the nuclides (s^{-1}), K_d is the distribution coefficient of the nuclides($m^3 g^{-1}$), ρ_s is the concentration of the suspended materials (g m^{-3}) and w_s is the settling velocity of suspended materials (m s^{-1}).

4.2 Results and discussion

4.2.1 Horizontal distribution of ^{239,240}Pu concentrations

The ^{239,240}Pu concentrations in the North Pacific water column are simulated by annual mean field because of less effort of seasonal field to model simulation after the 1970s. The spatial distribution of surface ^{239,240}Pu in 1963 primarily reflects that of global fallout. The surface ^{239,240}Pu concentrations greatly decrease in the mid-latitude region of the North Pacific. Relatively high ^{239,240}Pu concentration area in the sub-arctic and equatorial Pacific still appears in 1973, 1983 and 1993. In contrast of homogeneous surface ^{137}Cs concentrations in 1993, surface ^{239,240}Pu concentrations are high in the sub-arctic and equatorial Pacific and low in the subtropical Pacific till 1993. Evaluation of present ^{239,240}Pu concentrations in the North Pacific based on the HAM database suggests that surface ^{239,240}Pu concentrations are high in the sub-arctic and equatorial Pacific and low in the subtropical Pacific (Hirose and Aoyama, submitted). Therefore, this model calculation can apparently reproduce the geographic distributions of surface ^{239,240}Pu in the North Pacific. Causes of higher ^{239,240}Pu concentrations in the equatorial Pacific may be due to the upwelling to reduce the scavenging effect, whereas causes in sub-arctic Pacific is still unknown.

4.2.2 Vertical distribution of ^{239,240}Pu concentrations

The vertical distributions of the observed ^{239,240}Pu concentrations in each decade from the 1960s to the 1990s in the same area in Section 3.2.2 of ^{137}Cs. In the 1960s, there are no vertical profiles and a few surface ^{239,240}Pu concentrations in the HAM database. The model suggests that the most of the ^{239,240}Pu in the water column in the 1960s exists in surface waters and have no subsurface maximum. In the 1970s, 1980s and 1990s, the model calculation well reproduces the mid-depth maximum of the ^{239,240}Pu concentrations in water column, which is a typical vertical profile of plutonium observed in seawater (Bowen *et al.*, 1980; Nagaya and Nakamura, 1984; Livingston *et al.*, 2001). However, calculated surface ^{239,240}Pu concentrations underestimates in 1990s. This suggests that the penetration of ^{239,240}Pu from surface to subsurface might be faster than that in actual process in this model. Another important feature of observed vertical ^{239,240}Pu profiles is that ^{239,240}Pu maximum moves downward with time (Livingston *et al.*, 2001). This model succeeds to reproduce the downward movement of ^{239,240}Pu maximum by considering scavenging.

5. Future plan

In future, we plan online calculation for ^{137}Cs and ^{239,240}Pu by considering inter-annual variability.

At fitst, using a ealistically forced, global 3-D numerical ocean general circulation model (OGCM), we examine how well CFC tracer age approximates "ideal age" in the North Pacific thermocline as a function of time and explore the magnitude of interannual variability in both age measures. The CFC and ideal age fields are calculated using the Parallel Ocean Program with the KPP vertical mixing scheme and the Gent-McWilliams eddy parameterization. The horizontal resolution is approximately 1 degree, with 40 levels in the vertical. Surface momentum, heat and freshwater fluxes were computed using a bulk flux forcing scheme from NCEP reanalysis data from 1958 to 2000 (winds, air temperature, humidity) and satellite data products (clouds, insolation, precipitation).

The CFC tracer age distributions are computed on isopycnal surfaces through the main thermocline using the partial pressure and ratio approach. We focused on the shallow layer in the North Pacific where the ages are less than 30 years. The CFC tracer age agrees broadly with the simulated ideal age fields in the subpolar but tends to underpredict ideal age in the lower thermocline and tropics. These deviations are expected due to non-linear mixing effects where the mean ideal age values approach the length of the CFC transient. CFC tracer age increased from 1980s on the whole North Pacific, on the other hand, ideal age did not increase significantly. This suggests that increase of CFC tracer age in this model is caused not only by decrease of intermediate water mass formation but also by non-linear mixing effect after 1980s,

Acknowledgement

We are grateful to Frank Bryan for providing the model data and his helpful comments on the manuscript. We wish to thank Scott Doney and Matthew Hecht for providing their model data and their helpful comments. We thank Gideon Henderson and Christoph Heinze for their constructive comments on an earlier version of the manuscript. We thank Koki Maruyama and Norikazu Nakashiki for their helpful discussions; and Fukiko Taguchi for her technical help. This work was partly founded by former Science and Technology Agency, Japan.

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