Optimization of the horizontal shape of CO₂ injected domain and the depths of release in moving-ship type CO₂ ocean sequestration

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Abstract In moving-ship type CO₂ ocean sequestration, liquid CO₂ is discharged into a domain in a water column. Since the maximum CO₂ concentration that is reached depends on the horizontal shape of the water column and the depths of release, it is very important to optimize these parameters for each injection site in order to minimize the biological impact. We conducted numerical experiments using an offline Oceanic General Circulation Model with a horizontal resolution of 0.1 degree × 0.1 degree. Experiments using a different horizontal site shape show that a site elongated in the meridional direction is effective to reduce the CO₂ concentration. This is because CO₂ has a tendency to be transported in a zonal direction. Optimization of the vertical distribution of CO₂ injections is inherently determined by the balance of the following two factors; (1) dilution effect by eddy activity which decreases with depth, and the (2) predicted no effect concentration (PNEC), a criterion concentration causing no effect on biota, which increases with depth. Based on superposition of simulated CO₂ concentration, we determined the optimized vertical distribution of CO₂ injection which keeps

the ratio of a simulated maximum CO₂ concentration to PNEC constant.

Keywords CO₂ ocean sequestration · Carbon dioxide capture and storage (CCS) · Injection site · Biological impacts · PNEC

1 Introduction

CO₂ ocean sequestration has been proposed as an effective strategy to mitigate global warming [1]. In the moving ship method [2, 3], liquid CO₂ is discharged into the mid-depth ocean from a suspended conduit. Figure 1 presents a sketch of the moving ship concept. CO₂ is discharged within a site which is a domain in a water column within a few degrees of the horizontal. The released CO₂ forms droplets that slightly ascend due to buoyancy and dissolve into the surrounding water [4–6]. CO₂-enriched seawater is advected by currents from the injection site and diffuses into the surrounding water [7–9]. If the CO₂ concentration is decreased sufficiently by dilution, then the biological impacts are expected to be small. At time scales greater than a few months, related biological impacts are not acute but chronic. In an earlier numerical study, consecutive CO₂ injections were found to cause an increase in the CO₂ concentration during the first several to 10 years which eventually reaches an upper limit [10]. A biological study proposed an index that identifies no effect on biota, which is called predicted no effect concentration (PNEC) [11]. Results of many biological experiments [12] are used to estimate PNEC. The PNEC is defined as the highest CO₂ concentration which causes no effects on the weakest species divided by assessment factors with various uncertainties taken into

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Fig. 1 Concept of a CO₂ injected domain (site), where ship size is exaggerated

account [13]. Note that PNEC is a tentative standard and it should be further refined as additional data of biological experiments are obtained. To satisfy the condition that the simulated maximum CO₂ concentration is less than PNEC, the maximum CO₂ injection flux was determined by simulations to be 28 Mton/year for an injection site of size 1 degree $\times$ 1 degree in horizontal (131.4°E–132.4°E, 22.0°N–23.0°N, 977–2012 m) [10] and 62.5 Mton/year for another site of size 3 degrees in latitude and 1 degree in longitude (133.0°E–134.0°E, 19.0°N–22.0°N, 977–2012 m) [14].

Since the maximum CO₂ concentration that is reached depends on the lateral boundaries of injection (horizontal shape) from a moving ship and the depths of release, it is very important to optimize these parameters for each site. Although the specific location is also important, this has already been explored in a previous study [15]. Doubling the injection site area does not necessarily allow us to inject twice the amount of CO₂, since the maximum CO₂ concentration will become higher. Between two sites which have same the volume and have different horizontal shapes, the maximum CO₂ concentrations will be different under the same conditions of CO₂ injection flux. In a previous numerical study, where CO₂ is injected in some sites of 1 degree $\times$ 1 degree, we found that most of the CO₂-dissolved water is transported in a zonal direction [15]. It is known that the combination of Rossby waves and turbulence can lead to the formation of zonal velocity [16]; this might be related to the zonal CO₂ transport. Therefore, we expected that a site extended in the meridional direction would dilute CO₂ effectively, and tried to confirm the prospects in the paper. The first objective of the paper is to investigate how the maximum CO₂ concentration depends on the horizontal shape of a site. We conducted numerical experiments in which CO₂-injected sites have different horizontal shapes.

The second objective is to optimize the vertical distribution of injected CO₂. Initial vertical distribution of injected CO₂ is determined by injection depths and sizes of droplets. Therefore, to optimize the vertical distribution gives important information regarding injection depths and sizes of droplets in order to avoid chronic impacts on biota. In previous numerical studies where the vertical distributions of injected CO₂ are pre-determined, the maximum CO₂ concentration is close to PNEC around 1700 m while it is far smaller than PNEC at some other depths [14]. The objective of the optimization is to keep the ratio of the maximum CO₂ concentration to PNEC constant. If so, we can inject the maximum amount of CO₂ as the maximum concentration correspondent with PNEC, or the maximum concentrations have the same margin to PNEC at all depths. The optimized vertical distribution of injected CO₂ is considered to be determined by the balance of the following two factors. PNEC increases with depth where CO₂ injection is implemented. However, eddy activity which works to effectively dilute CO₂ becomes smaller with depth increasing [10].

2 Model and design of experiments

2.1 Model

CO₂ concentration is calculated by an offline model with a horizontal resolution of 0.1 degree $\times$ 0.1 degree. The offline model is based on the Coupled Ocean-Sea-Ice Model for the Earth Simulator (OIFES) developed at the Earth Simulator Center/Japan Agency for Marine-Earth Science and Technology (JAMSTEC) [17, 18]. The model has 54 vertical layers with realistic geometry. Horizontal and vertical velocities are not calculated in the offline model but daily-mean velocities calculated in OIFES (online model) applied in the North Pacific are used. We conducted a simulation for 30 years and the daily-mean velocities during 5 years are repeated six times. The integration during 30 years is considered to be sufficient long to simulate the maximum concentration of CO₂, since previous numerical results showed that CO₂ concentration nearly reaches its upper limit within 10 years from the beginning of the injection [10]. The domain of the offline model is 120°E–180°E, 10°N–50°N. Since the model has open lateral boundaries, the total amount of the injected CO₂ is not accurately conserved. However, a previous study using the online model applied in the North Pacific (98°E–70°W, 20°S–68°N) confirmed that almost all of the injected CO₂ remains in the domain for 30 years if CO₂ is injected in Japanese waters [15].

We treated CO₂ as a purely passive tracer dissolved in seawater. Since we simulated CO₂ transport and dilution on

Table 1 Design of experiments

Exp.	Site location	Injection depth (m)	Injection flux (Mton/year)	Injection rate per volume (ppm/year)
H1x3	133°E–134°E, 19°N–22°N	977–2719	50	0.077–1.45
H3x1	132°E–135°E, 20°N–21°N	977–2719	50	0.077–1.45
H1x2	133°E–134°E, 19.5°N–21.5°N	977–2719	50	0.116–2.18
H1x1_2	133°E–134°E, 19°N–20°N 133°E–134°E, 21°N–22°N	977–2719	50	0.116–2.18
V1	133°E–134°E, 19°N–22°N	977–1113	4.86	1.0
V2	133°E–134°E, 19°N–22°N	1113–1264	5.37	1.0
V3	133°E–134°E, 19°N–22°N	1264–1429	5.88	1.0
V4	133°E–134°E, 19°N–22°N	1429–1609	6.40	1.0
V5	133°E–134°E, 19°N–22°N	1609–1803	6.91	1.0
V6	133°E–134°E, 19°N–22°N	1803–2012	7.42	1.0
V7	133°E–134°E, 19°N–22°N	2012–2234	7.92	1.0
V8	133°E–134°E, 19°N–22°N	2234–2470	8.40	1.0

a time scale longer than several weeks after the initial dilution processes are completed, including dissolution of droplets and plume dynamics, we assumed that the CO_2 concentration is low enough not to cause vertical movement due to density differences. A liquid droplet-plume model showed that a vertical movement of seawater due to density difference does not occur if the CO_2 concentration is lower than $0.02\text{--}0.03\text{ kg/m}^3$ [4]. Since the maximum CO_2 concentration is estimated to be about 0.002 kg/m^3 in our previous study [14], vertical movement does not occur. Our model does not express subgrid phenomena, which would be potential sources of error. We cannot directly validate our simulation for ocean sequestration, but it is encouraging that the simulation of chlorofluorocarbon using the same model agrees well with the observations [19].

2.2 Experiments to optimize the horizontal shape of a site

To assess effects of the horizontal shape of a site on CO_2 concentration, three CO_2 injection experiments were conducted in addition to Exp. H1x3 in a previous study [14], in which CO_2 is injected in a site of 1 degree in longitude and 3 degrees in latitude (Table 1). The ocean area around the site has been studied for many years by the research project for CO_2 sequestration managed by the Research Institute of Innovative Technology for the Earth, and physical, chemical and biological data have been accumulated. In Exp. H3x1 and H1x2, we used sites of 3 degrees in longitude and 1 degree in latitude, and 1 degree in longitude and 2 degrees in latitude, respectively. In Exp. H1x1_2, CO_2 is injected simultaneously into two sites of $1\text{ degree} \times 1\text{ degree}$ at a 1 degree

Fig. 2 Vertical distribution of injected CO_2 in Exp. H3x1, H1x2, and H1x1_2

interval of latitude. By the experiments, effects of site area and shape on CO_2 concentration are examined. Comparing H1x3 and H3x1, we examine the concept that a site extended in latitude is more effective to dilute the CO_2 concentration. In all the experiments, CO_2 is consecutively injected at a rate of 50 Mton/year. Note that CO_2 injection rate per unit volume is larger in H1x2 and H1x1_2 than in H1x3 and H3x1, where 50 Mton/year CO_2 is injected into the area of 2 degrees^2 in the formers and 3 degrees^2 in the latter. While CO_2 injection is uniform on a horizontal plane, it depends on depth. The

vertical distribution of the injected CO₂ is shown in Fig. 2 [20]. This is obtained based on results of two liquid droplet models [21, 22] under the following assumptions: the same amount of CO₂ is injected from pipes at depths of 2500, 2290, 2080, 1970, 1830 and 1640 m, and CO₂ droplet size is 14 mm.

2.3 Experiments to optimize vertical distribution of injected-CO₂ amount

Our model has eight layers in 977–2470 m in which CO₂ is assumed to be injected. Corresponding to the eight layers, we conducted eight experiments V1–V8 in which CO₂ is injected into only one layer. Horizontal shape of a site is the same as that of H1x3, since the shape of H1x3 has an advantage to dilute CO₂ effectively, as shown later. CO₂ injection flux per unit volume is 1.0 ppm/year, which is common in V1–V8. Since the CO₂ equation is linear, superposition of the simulated CO₂ concentrations multiplied by a constant also satisfies the equation for CO₂

concentration. If a set of the constants is properly selected, we can keep the ratio of a maximum CO₂ concentration to PNEC constant through the CO₂-injected layers from 977 to 2470 m.

3 Results

3.1 Experiments to optimize the horizontal shape of a site

After 30 years from the beginning of the CO₂ injection, CO₂ extends a thousand kilometers in H1x3, H3x1, H1x2 and H1x1_2 (Fig. 3). Most of the CO₂-dissolved water is transported in a zonal direction, as in the previous study [14]. Relatively high CO₂ concentration is obtained around the injection sites. The maximum concentration at 1904 m is higher in H1x2 and H3x1 than in H1x3 and H1x1_2. For each model layer, we calculated the maximum CO₂ concentration during 30 years, and compared it to PNEC

Fig. 3 Horizontal distributions of monthly mean CO₂ concentration at 1904 m after 30 years from the beginning of the injection in Exp. **a** H1x3, **b** H3x1, **c** H1x2, and **d** H1x1_2

Fig. 4 Maximum concentration (ppm) during 30 years at each model layer in Exp. H1x3 (solid thin line), H3x1 (dashed and dotted line), H1x2 (dotted line) and H1x1_2 (dashed line). PNEC is shown by the thick solid line

(Fig. 4). PNEC in ppm (CO_2 mass/seawater mass) was converted from PNEC of $\Delta 500 \mu\text{atm}$ [11, 20], where “ Δ ” means additional CO_2 to the background CO_2 . In the conversion, physical and chemical quantities necessary in calculating the chemical reaction of CO_2 , for example, temperature, alkalinity, are based on ship observation around 132°E , 22.5°N , which was carried out as a part of the research project for CO_2 ocean sequestration. PNEC in ppm is the lowest around 900 m, since the background CO_2 concentration is high around the depth. The simulated maximum concentration is higher in the order of H1x2, H3x1, H1x1_2 and H1x3. Although the CO_2 injection flux has a peak at 1702 m, the maximum concentration is obtained at 1904 m except for H1x1_2. This is due to the dispersion by eddies decreases with depth. Comparing H1x3 and H3x1, we confirmed our expectation that a site extended in latitude is effective to dilute CO_2 concentration. CO_2 injection flux per volume is about 1.5 times larger in H1x2 and H1x1_2 than in H1x3 and H3x1. In H1x2, the highest concentration in the four experiments was obtained at 1904 m, which is higher than PNEC. However, the vertical distribution of the maximum CO_2 concentration in H1x1_2 is nearly equal to that in H1x3. The comparison of H1x2 and H1x1_2 suggests that CO_2 injection sites in a meridional interval are effective to dilute CO_2 . In H1x1_2 (H1x2), if we consider that CO_2 is separately injected into the northern and southern half area of $1^\circ \times 1^\circ$, the overlap of the resulted two sets of CO_2 distribution determines the maximum concentration. The smaller maximum in H1x1_2 than in H1x2

would be resulting from the smaller amount of the overlap in H1x1_2 than in H1x2. The maximum CO_2 concentration in H1x1_2 is slightly higher than that in H1x3, and the maximum in H1x1_2 is achieved at a depth where PNEC is relatively small. As a result, the ratio of the maximum concentration to PNEC is larger in H1x1_2 than in H1x3. Therefore, we conclude that the site in H1x3 is the optimal horizontal shape of the four experiments.

3.2 Experiments to optimize the vertical distribution of injected- CO_2

Vertical distribution of injected CO_2 was optimized, using the results of the experiments V1–V8. The distributions of CO_2 concentration at CO_2 -injected depth after 30 years from the beginning of the injection are shown in V1, V4, V6 and V8 (Fig. 5). CO_2 concentration increases with depth, since dilution of CO_2 by mesoscale eddies decreases with depth. Relatively large CO_2 concentrations occur around the injection sites. We calculated the maximum concentration for each model layer in every experiment (Fig. 6). The maximum concentrations at different layers do not necessarily occur simultaneously. The maximum CO_2 concentration increases as CO_2 -injected depth increases, where CO_2 injection flux per unit volume is the same in V1–V8. The distribution of the maximum concentrations takes a peak at the CO_2 -injected layer. In many cases, the CO_2 concentration in layers below the CO_2 -injected layer is larger than that in upper layers, where the CO_2 concentration in layers below the CO_2 -injected layer is caused by vertical advection and diffusion of CO_2 -enriched seawater.

To optimize the vertical injection amount, we considered the following equation:

$$\begin{pmatrix} a_{V1,1} & a_{V2,1} & \cdots & a_{V8,1} \\ a_{V1,2} & a_{V2,2} & \cdots & a_{V8,2} \\ \vdots & \vdots & \ddots & \vdots \\ a_{V1,8} & a_{V2,8} & \cdots & a_{V8,8} \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ \vdots \\ x_8 \end{pmatrix} = \begin{pmatrix} c_1 \\ c_2 \\ \vdots \\ c_8 \end{pmatrix}, \quad (1)$$

where $a_{i,j}$ shows the maximum concentration obtained in Fig. 6a, and the first and second indexes denote the experiments and model layer, respectively. For example, $a_{V1,1}$ is the maximum concentration in V1 at the uppermost layer (977–1113 m) in the CO_2 -injected layers in V1–V8. In the equation, c_1 – c_8 show PNEC at the model layers, and x_1 – x_8 are coefficients to be solved, which give an optimized CO_2 injection flux. If the coefficients are properly given, we can keep the ratio of maximum concentration to PNEC 1.0 through all the model layers in which CO_2 is injected in V1–V8. The CO_2 concentration above (<977 m) and below (>2470 m) the CO_2 -injected layer are small and are ignored in the estimation. Note that this is a conservative

Fig. 5 Horizontal distributions of monthly mean CO₂ concentration at CO₂-injected depth after 30 years from the beginning of the injection in Exp. **a** V1 (1041 m), **b** V4 (1515 m), **c** V6 (1904 m), and **d** V8 (2349 m)

estimation since the maximum concentration $a_{i,j}$ in Eq. 1 do not necessarily occur simultaneously. Since the Eq. 1 is linear, the following equation is also obtained;

$$\begin{pmatrix} a_{V1,1} & a_{V2,1} & \cdots & a_{V8,1} \\ a_{V1,2} & a_{V2,2} & \cdots & a_{V8,2} \\ \vdots & \vdots & \ddots & \vdots \\ a_{V1,8} & a_{V2,8} & \cdots & a_{V8,8} \end{pmatrix} \begin{pmatrix} \alpha x_1 \\ \alpha x_2 \\ \vdots \\ \alpha x_8 \end{pmatrix} = \begin{pmatrix} \alpha c_1 \\ \alpha c_2 \\ \vdots \\ \alpha c_8 \end{pmatrix}, \quad (2)$$

where α is the reciprocal of a safety factor. The positive constant α takes a value less than 1.0. Once x_1 – x_8 are calculated, corresponding to an arbitrary safety factor we can easily estimate the CO₂ injection flux which keeps the ratio of maximum concentration to PNEC α through the eight model layers.

The calculated x_1 – x_8 values are shown in Fig. 7. At the each of the model layers, if we inject a CO₂ flux of 1.0 ppm/year times the coefficients x_j , the estimated maximum CO₂ concentration will be the same as PNEC. Without causing CO₂ larger than PNEC, up to 86.7 Mton CO₂/year $\left(\sum_{j=1}^8 x_j V_j \rho\right)$ can be injected, where V_j is the

volume of the each model layer, and ρ is seawater density. Note that this value will be revised if PNEC is refined based on results of future biological experiments. We can inject relatively larger CO₂ in the upper layer than in the deeper layer without causing CO₂ concentration larger than PNEC. As shown above, PNEC increases with depth where CO₂ injection is implemented, but eddy activity diluting CO₂ concentration becomes small with depth. In the optimization of CO₂ injection flux, the effect of the latter is larger than that of the former.

4 Summary and discussion

In moving-ship type CO₂ ocean sequestration, liquid CO₂ is discharged into a domain in a water column. Since the maximum CO₂ concentration that is reached depends on the horizontal shape of the water column and the depths of release, it is very important to optimize these parameters for each injection site in order to minimize biological impact. To simulate CO₂ concentration, we used an offline

Fig. 6 Maximum CO₂ concentration at each layer simulated in Exp. **a** V1, **b** V2, **c** V3, **e** V4, **f** V5, **g**, V6, and **h** V8. The maximum concentration is calculated within 1 degree from the site (132°E–135°E, 18°N–23°N)

model based on OIFES with a horizontal resolution of 0.1 degree $\times$ 0.1 degree. Simulated maximum CO₂ concentration is compared with PNEC which is an index causing no effect on biota. To optimize the horizontal shape of an injection site, we conducted experiments in which CO₂-injected sites have a different horizontal shape. Comparing the two experiments in which the injection sites are of 1(lon) $\times$ 3(lat) and 3(lon) $\times$ 1(lat) degree, we showed that a site extended in a meridional direction is effective to

dilute injected CO₂. This is because CO₂ has a tendency to be transported in a zonal direction. The experiment with the two sites at 1 degree intervals in latitude showed that the meridional interval is effective to decrease CO₂ concentration. The experiment shows the possibility that we can set some sites around Japan without increasing the concentration of CO₂ over PNEC. If we can set multiple sites without causing biological impacts, it enables us to sequester huge amounts of CO₂ into the ocean.

Fig. 7 Optimized vertical distribution of the injected CO₂ amount, which gives the same ratio of the maximum CO₂ concentration to PNEC in all CO₂-injected layers. In this case, the ratio is 1

To optimize the vertical distribution of CO₂ injection, we conducted eight experiments in which CO₂ is injected into only one layer of eight layers from 977 to 2470 m. Since the equation for CO₂ concentration is linear, superposition of the simulated CO₂ concentrations multiplied by a constant also satisfies the equation for CO₂ concentration. We estimated the constants which keep the ratio of a maximum CO₂ concentration to PNEC constant through the CO₂-injected layers. The estimated constants which show possible injection flux without biological impacts are relatively larger CO₂ in upper layers than in deeper layers. In the two important factors affecting biological impacts, PNEC increases with depth where CO₂ injection is implemented, but eddy activity diluting CO₂ concentration becomes small with depth. The result of the estimation shows that the effect of the latter is larger than the former. In a real ship operation, vertical distribution of CO₂ injection flux results from dissolution of CO₂ liquid droplets into the surrounding water. We need to design the proper length of injection pipes, CO₂ droplet sizes and CO₂ injection flux from pipes, as the resultant vertical distribution of CO₂-injection flux is close to that estimated in the paper. This is beyond the scope of the paper and will be explored by a modeling team using liquid-droplet models in future.

In the above optimization, we focused on one site located southeast of Japan. Optimization in other sites is discussed below. A previous study [15] in which CO₂ is injected in several sites in different ocean areas showed that most of the CO₂-dissolved water is transported in the zonal direction. Therefore, a site extended in a zonal

direction would be effective to dilute CO₂ concentration in a wide ocean area. Optimized vertical distribution of injection flux will be considerably different among sites, since eddy activity differs by a factor of 10 in different ocean areas [15]. However, the optimizing method presented in the paper is also applicable.

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