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Response to Comment on "Fate of Rising CO_2 Droplets in Seawater"

I welcome the comment by Alendal et al. (*1*), and the opportunity to clarify some issues. My paper (*2*) focuses on theoretical calculation of the dissolution rate of a single rising CO_2 droplet (or noninteracting droplets) in the context of carbon sequestration in oceans. The calculated dissolution rate agrees with the in situ experimental data of Brewer et al. (*3*). The comment (*1*) does not raise any issue on the fundamental theoretical treatment. My paper (*2*) also treated a few other issues but in less detail. Alendal et al. (*1*) raise four points, mostly on these other issues, or on issues not treated by my paper (*2*), such as biological impact. The points are discussed below in sequence.

1. The possibility for injected CO_2 to induce an eruption similar to lake eruptions. Because rising CO_2 droplets injected into ocean water would convert to gas bubbles at shallow depth (about 400 m but depending on temperature), I (*2*) suggested that to "avoid the formation of rising CO_2 droplet plumes and to avoid CO_2-driven eruptions, injection of CO_2 liquid into the shallow oceans needs to be controlled, using low injection rates and dispersing CO_2 liquid to a large volume of seawater. Safer injection schemes include injection of CO_2 liquid to great enough depths that liquid CO_2 density is greater than ambient seawater so that CO_2 liquid would sink and gradually dissolve in seawater." Alendal et al. (*1*) quoted the IPCC report (*4*) that "there in no known mechanism that could produce an unstable volume of water containing 2 $MtCO_2$ at depth shallower than 500 m, and thus no mechanism known by which ocean storage could produce a disaster like that at Lake Nyos".

The IPCC report (*4*) is an authoritative document on carbon storage. Nonetheless, details of the IPCC report are not necessarily accurate and quoting the IPCC report does not necessarily end a debate. For example, the above quotation of the IPCC report cites two million tons of CO_2 as the amount released in the 1986 eruption of Lake Nyos, likely based on Kling et al. (*5*) (although another reference was cited, which does not contain the information and might be a typographical error). However, the issue was revisited after the work of Kling et al. (*5*) and consensus was reached that the amount of CO_2 erupted was about 15% of the above quoted value (*6*). Furthermore, such an amount of CO_2 is not necessary for a CO_2-driven eruption; the amount of CO_2 released by the 1984 Lake Monoun eruption was much less.

One may envision both types of scenarios: (i) slow injection of individual droplets would not induce an eruption, and (ii) as the IPCC report pointed out "if somehow large volumes of liquid CO_2 were suddenly transported above the liquid−gas-phase boundary, there is a possibility of a self-accelerating regime of fluid motion that could lead to rapid degassing at the surface" (*4*, p 308). The possibility and dynamics of ocean eruptions powered by the sudden formation of many bubbles have been discussed (*7, 8*). The threshold between the two extremes has not been mapped out. Contrary to the claim of Alendal et al. (*1*), at depth shallower than 2000 m, the plume water would be less dense if CO_2 is in the form of droplets or bubbles instead of the dissolved form.

I maintain that (i) new research is necessary for understanding the condition under which safe injection can be achieved, and (ii) it is necessary to exercise caution in injecting a large amount of CO_2 into the deep ocean. As long as the injection design is carefully evaluated, escape of CO_2 to the surface through bubble plumes is avoidable. Claiming that such caution is unnecessary would not be wise.

2. Modeling of terminal velocity. I (*2*) focused on new developments and did not repeat previous calculations. Because Brewer et al. (*3*) already modeled terminal velocity well using a simple formulation, I (*2*) did not present results of new calculations of terminal velocity. Furthermore, to model the reported rising velocity accurately, it is necessary to know (i) the in situ density of seawater, which was not reported by Brewer et al. (*3*); and (ii) the effect of the motion of the rising box of the remotely controlled vehicle on the overall ascent velocity of the two droplets, which would affect the measured rising velocity but would not significantly affect the dissolution rate. Gangstø et al. (*9*) tried to model the droplet ascent velocity data (*3*) and found that (i) the standard drag cannot model the data, (ii) a recent theory for the drag coefficient of bubbles (not shelled droplets) agrees with the data better, but (iii) no model can predict the observed high velocity at radius <1 mm. It seems that either a new theory is necessary or there is additional effect on the rising velocity.

The effect of the terminal rising velocity on the dissolution rate is not so major. For example, if the ascent velocity increases by 50%, the dissolution rate varies by 22%. Hence a more accurate terminal velocity might change the calculated dissolution somewhat, making the calculation either in slightly better agreement, or in slightly worse agreement, with data. Even if the agreement between calculation and data became slightly worse than that shown in ref *2*, the ability to calculate droplet dissolution rate without any adjustable parameter to an accuracy of about 20% by the method in ref *2* is still a significant advance. For comparison, a recent dissolution model (*9*) still contains an adjustable factor.

3. Dissolution from a lake of liquid CO_2 in ocean water. In addition to the main focus of dissolution rate of a rising CO_2 droplet, I (*2*) also calculated the dissolution rate of a CO_2 liquid lake with hydrate shell to be 0.35 m/yr, based on a formula in ref *10*, which is extracted from an established although approximate method (*11*). The model result of Fer and Haugan (*12*) is 0.1 m/yr, and that of Haugan and Alendal (*13*) is 0.63 m/yr (average of 5 simulations for flow velocity of 0.05 m/s). Without experimental data for direct comparison, it is not yet known which model matches reality best.

4. Whether new theory is needed. The last paragraph of Alendal et al. (*1*) seems to suggest that theoretical development is not useful; they wrote that "while we welcome new model approaches for single droplets, we suggest that the primary need is for more in situ experiments, including, particularly, studies of biological impact of elevated CO_2 levels". My paper (*2*) focused on the dynamic and kinetic fate of injected CO_2 droplets, and did not evaluate the biological impact, which of course does not mean that it is unimportant. Back to the question of the fate of injected CO_2 liquid in deep ocean, I maintain that both experimental data and theory that can predict (not just simply fit) data are needed because experiments can ground truth theory but cannot examine all oceanic conditions, whereas the theory in my paper (*2*) can be applied to any temperature-depth condition. (There are many examples of the extreme importance of theory in science, which do not need to be elaborated here.) I (*2*) focused only on the theory of the dissolution of a single rising droplet, or noninteracting droplets. Further theoretical development is necessary to investigate the fate of many CO_2 droplets interacting with

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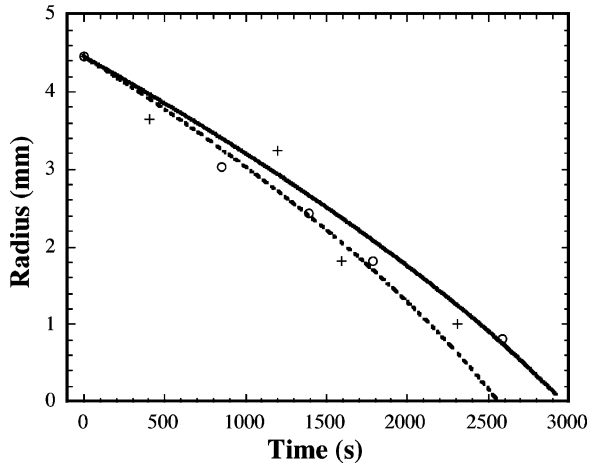

FIGURE 1. Corrected version of Figure 2 of Zhang (*2*). Points are experimental data for CO_2 droplet dissolution (*3*) and curves are calculated droplet dissolution as a function of time (*2*).

one another, and the threshold for the development of a CO_2 plume.

In summary, I would like to emphasize that the comment by Alendal et al. (*1*) did not challenge the basic theory of my model, but raised questions on some side issues. I maintain that if CO_2 is injected on large scale to a depth where CO_2 liquid density is less than that of seawater, it is prudent to understand the threshold injection rate for CO_2 plume eruption and then design the injection rate below the threshold. I concur with Alendal et al. (*1*) that many other aspects such as the biological impact of elevated CO_2 levels also need to be addressed.

Finally, unrelated to the comment, I would like to take this opportunity to make a correction of Figure 2 in Zhang (*2*). One data point was incorrectly plotted in the figure. The corrected version is shown in Figure 1. The correction does not significantly change the match between experimental data and my theoretical calculation.

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