

Comment on "Fate of Rising CO₂ Droplets in Seawater"

In a recent manuscript published in *Environmental Science and Technology* (1), Zhang warns about a possible Lake Nyos effect from CO₂ injection in the ocean. This analogy is not new and has been discussed earlier. Based on material that had been published and available prior to submission of Zhang's manuscript, the recent IPCC special report on Carbon Dioxide Capture and Storage [2, released December 2005] concluded: "Thus, there is no known mechanism that could produce an unstable volume of water containing 2 million tCO₂ 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 and some other material referenced herein were published after Zhang's article appeared in *ES&T*, and were therefore unavailable to him during preparation of the manuscript. But, his study does not introduce any new mechanism that could put large amounts of CO₂ in a buoyant state, and hence brings no new information that could alter the IPCC conclusion. In fact, his study only considers single droplets. In a real injection there would be a cloud of droplets that forms a buoyant plume, with completely different dynamics, that Zhang's simulations do not consider. The plume water would become denser, due to increased CO₂ concentration, and tend to sink out reducing the net vertical velocity of the droplets. In the confined freshwater Lake Nyos the water masses were saturated with CO₂. Even in the core of a droplet plume the seawater would be far from saturated (3–12, to mention some). Furthermore, the ocean would continue to dilute the concentration and ultimately the injected CO₂ would be transported by the oceanic currents as a passive tracer (13).

Zhang introduces a model for calculating the dissolution of CO₂ from the droplets, and validates it using a single-droplet model that he compares with the experiments by Brewer et al (14). However, it seems like his study contains several inaccuracies. The terminal velocity and the rate of dissolution from droplets are highly connected, but Zhang only considers the latter when comparing model and experimental data. One would expect droplets, under the assumptions described by Zhang, to rise 50% faster than observed. In fact, a constant drag coefficient equal to one, as used in the rough model by Brewer et al., gives a better fit on terminal velocity than the drag coefficient used by Zhang, at least in the initial phase of the droplet ascent (15). Some of this inaccuracy might be counteracted by another: the use of potential density as opposed to in situ density.

Zhang also comments upon the dissolution from a lake of liquid CO₂ on the sea floor. Elsewhere, this has been modeled by combining a capillary permeation model of the hydrate layer with various representations of turbulence in the ocean above (16, 17, see also relevant measurements by Rehder et al. in 18). Zhang's expression does not account for the applied external shear stress and local turbulence that is expected to alter the dissolution rate.

We recognize the need for further research on the behavior of CO₂ in oceanic waters. The single droplet experiments by Brewer et al. are the only in situ experiments yet performed.

Hence, so far all studies have had to rely on tank experiments (8, 10, 11), one single droplet in situ experiment, and theoretical considerations (19). 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 CO₂ levels (20). And most importantly, we maintain that there are well understood physical processes included in published model studies which demonstrate that there is no reason to expect CO₂-driven eruptions from CO₂ droplet releases in the open ocean.

Literature Cited

- (1) Zhang, Y. Fate of Rising CO₂ Droplets in Seawater. *Environ. Sci. Technol.* **2005**, *39*, 7719–7724.
- (2) IPCC. *Special Report on Carbon Dioxide Capture and Storage*; Prepared by Working Group III of the Intergovernmental Panel on Climate Change; Metz, B., Davidson, O., de Coninck, H. C., Loos, M., Meyer, L. A., Eds.; Cambridge University Press: Cambridge, U.K., 2005; <http://www.ipcc.ch/activity/srccs>.
- (3) Hirai, S.; Okazaki, K.; Tabe, Y.; Hijikata, K.; Mori, Y. Dissolution rate of liquid CO₂ in pressurized water flows and the effect of clathrate films. *Energy* **1997**, *22*, 285–293.
- (4) Adams, E. E.; Crounse, B. C.; Harrison, T. H.; Socolofsky, S. A. Analytical and experimental studies of droplet plumes with application to CO₂ ocean sequestration. *Abstr. Papers Am. Chem. Soc.* **2000**, *220*, U401–U401.
- (5) Alendal, G.; Drange, H. Two-phase, near-field modeling of purposely released CO₂ in the ocean. *J. Geophys. Res.* **2001**, *106*, 1085–1096.
- (6) Sato, K.; Sato, T. A study on bubble plume behavior in stratification. *J. Marine Sci. Technol.* **2001**, *6*, 59–69.
- (7) Sato, T.; Sato, K. Numerical prediction of the dilution process and its biological impacts on CO₂ ocean sequestration. *J. Marine Sci. Technol.* **2002**, *6*, 169–180.
- (8) Socolofsky, S. A.; Adams, E. E. Multi-phase plumes in uniform and stratified crossflow. *J. Hydraul. Eng.* **2002**, *40*, 661–672.
- (9) Chen, B.; Song, Y.; Nishio, M.; Akai, M. Large-eddy simulation of double-plume formation induced by CO₂ dissolution in the ocean. *Tellus* **2003**, *55B*, 723–730.
- (10) Socolofsky, S. A.; Adams, E. E. Liquid volume fluxes in stratified multiphase plumes. *J. Hydraul. Eng.* **2003**, *129*, 905–914.
- (11) Socolofsky, S. A.; Adams, E. E. Role of slip velocity in the behavior of stratified multiphase plumes. *J. Hydraul. Eng.* **2005**, *131*, 273–282.
- (12) Chen, B.; Song, Y.; Nishio, M.; Someya, S.; Akai, M. Modeling near-field dispersion from direct injection of carbon dioxide into the ocean. *J. Geophys. Res.* **2005**, *110*, C09S15; doi: 10.1029/2004JC002567.
- (13) Drange, H.; Alendal, G.; Johannessen, O. M. Ocean release of fossil fuel CO₂: A case study. *Geophys. Res. Lett.* **2001**, *29*, 2637–2640.
- (14) Brewer, P.; Peltzer, E.; Friederich, G.; Rehder, G. Experimental Determination of the Fate of Rising CO₂ Droplets in Seawater. *Environ. Sci. Technol.* **2002**, *36*, 5441–5446.
- (15) Gangstø, R.; Haugan, P. M.; Alendal, G. Parameterization of drag and dissolution of rising CO₂ drops in seawater. *Geophys. Res. Lett.* **2005**, *32*, L10612.
- (16) Fer, I.; Haugan, P. M. Dissolution from a liquid CO₂ lake disposed in the deep ocean. *Limnol. Oceanogr.* **2003**, *48*, 872–883.
- (17) Haugan, P. M.; Alendal, G. Turbulent diffusion and transport from a CO₂ lake in the deep ocean. *J. Geophys. Res.* **2005**, *110*, C09S14.
- (18) Rehder, G.; Kirby, S. H.; Durham, W. B.; Stern, L. A.; Peltzer, E. T.; Pinkston, J.; Brewer, P. G. Dissolution rates of pure methane hydrate and carbon-dioxide hydrate in unsaturated seawater at 1000-m depth. *Biochim. Cosmochim. Acta* **2004**, *68*, 285–292.

* Corresponding author e-mail: Guttorm.Alendal@bccs.uib.no.

- (19) Radhakrishnan, R.; Demurov, A.; Herzog, H.; Trout, B. L. A consistent and verifiable macroscopic model for the dissolution of liquid CO₂ in water under hydrate forming conditions. *Energy Conv. Mgmt.* **2003**, *44*, 773–782.
- (20) Orr, J. C.; Pantoja, S.; Pörtne, H.-O. Introduction to special section: The ocean in a high-CO₂ world. *J. Geophys. Res.* **2005**, *110*, C09S01; doi: 10.1029/2005JC003086.

Guttorm Alendal* and Peter M. Haugan

University of Bergen, Norway

Reidun Gangstø

Laboratoire des Sciences du Climat et de
L'Environnement, France

Ken Caldeira

Carnegie Institution of Washington

Eric Adams

Massachusetts Institute of Technology

Peter Brewer and Edward Peltzer

Monterey Bay Aquarium Research Institute

Gregor Rehder

Leibniz-Institut für Meereswissenschaften, Germany

Toru Sato

University of Tokyo, Japan

Baixin Chen

National Institute of Advanced Industrial Science and
Technology (AIST), Japan

ES052458C