Supporting Information

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"A kinetic model of gas bubble dissolution in groundwater and its implications for the dissolved gas composition"

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Sensitivity of the model to multiple bubble sizes

To examine the influence of a distribution of different bubble sizes on the dissolved concentrations, the illustrative model simulations were repeated employing now four different bubble size classes instead of only one single bubble size (0.25 mm, 0.3 mm, 0.35 mm, 0.4 mm). The other model parameters were not changed. The results are shown in Figure 1. From the bubble radii, we see that a redistribution of the bubble sizes occurs in both scenarios – the vertical flow and the stagnating situation. In both cases, a progression in the bubble extinction sequence exists which begins with the smallest bubbles and ends with the largest bubbles. While the smaller bubbles dissolve, the larger bubbles grow until finally the last remaining bubble size class is either dissolved completely (advective flow regime) or maintained stable at the new equilibrium with the surrounding water (stagnant flow regime). The reason for this characteristic behavior is the difference in the local dissolved equilibrium concentrations for bubbles of different size. Due to the higher capillary pressure in small bubbles, their local equilibrium concentrations in water are higher than for larger bubbles. Thus in a multi-bubble size system dissolved gas concentrations in water are reached that represent a saturation deficiency with respect to the smallest bubbles, forcing them to dissolve, but correspond to a local supersaturation with respect to the larger bubbles, forcing these bubbles to grow. As a result, the bubbles will be re-arranged in size, leaving only one final bubble class. For this specific bubble size the hydrostatic pressure plus the capillary pressure are balanced by the gas pressure inside the bubble. It can therefore be considered stable.

If the relative concentrations of the multiple bubble size simulations displayed in Figure 1b and 1d are compared to the results obtained by employing just one single bubble size, it is obvious that the general dynamics of the gas dissolution is nearly identical. The main difference are small concentration peaks evident in the multi-bubble size simulations that result from the extinction of the respective smaller bubble sizes. While the concentration evolution is similar in both cases, the time scale is prolonged in the multi-bubble size simulations, depending on the actual flow regime. Apparently, the re-distribution of the entrapped gas between different bubble sizes becomes the time controlling process considering no flow conditions. In this case, the bubble size rearrangement takes place only diffusively, and hence the time needed to reach the final equilibrium is prolonged by a factor of approximately 1400 (from minutes to days) compared to the single size simulation, although the major initial change in concentrations still occurs quickly. In the advective flow system, the time until all bubbles are dissolved is only slightly increased. Here, the advective transport of the dissolved gases continues to control the bubble dissolution process.

Figure Caption:

Figure 1. Bubble radii and relative outflow concentrations of He, Ne, Ar, Kr, Xe, N_2 and O_2 from the KBD-model simulation for 4 distinct bubble size classes. Fig. 1a and 1b show the results for stagnant, no-flow conditions. Fig. 1c and 1d display the simulations for an advective vertical downward water flow.

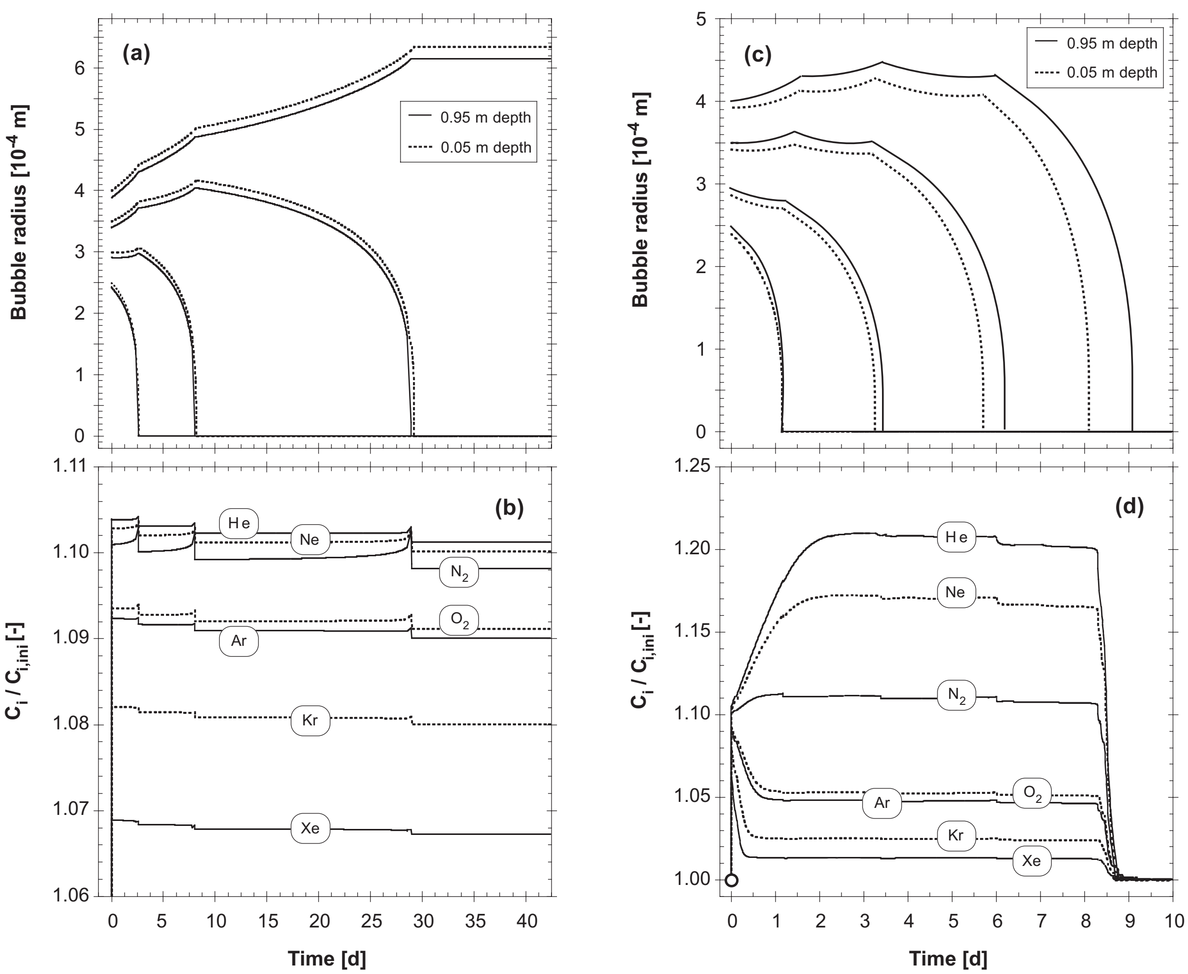

Figure 1 Supplement