Quantifying temperature-dependent methane ebullition from reservoirs via systems analysis

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Sediments accumulating high carbon loads in shallow water bodies such as lakes and reservoirs are potential hot-spots of methane emission via gas bubbles (ebullition). Here we show that a run-of-the river reservoir in Switzerland emitted >150 mg CH_4 m^{-2} d^{-1} during the warm season [1].

Direct measurements of ebullition rates suffer from enormous spatial and temporal variability. We therefore combined integrating observations such as the methane concentrations at the inflow and outflow with a mass balance approach. The analysis revealed a strong positive correlation between temperature and the accumulation of dissolved methane in the water column due to bubble dissolution. The accumulation rates in the water column increased by more than a factor of 3 when the water temperature changed from 10 to 17°C. Methane emission rates to the atmosphere were then estimated with the help of a bubble plume model which calculated the fraction of methane lost to the water column by raising bubbles.

This systems analysis was validated with direct observations of bubble fluxes by funnels and floating chambers. The approach is generally applicable for run-of-the river schemes showing an increase of methane concentrations along the flow path. The substantial methane emissions discovered in this case study suggest that high emission rates are not limited to tropical reservoirs and that temperature might act as a driver for high ebullition rates.

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Sulfur isotope systematics of degassing arc basalts: Implications from Masaya volcano

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Sulfur isotope fractionation during volcanic degassing and sulfate aerosol formation remains poorly constrained, despite the importance of these processes in the S cycle. We present S isotope compositions of scoria, magmatic gases, and sulfate aerosol from Masaya (MA) volcano (Nicaragua) in the first study to characterize all S phases involved in volcanic degassing to the atmosphere.

The $\delta^{34}S_{V-CDT}$ value of scoria from MA is ~+7.9‰, while that of SO_2 collected from the plume using flow-through bubblers is +5.0±0.7‰ (n=7). No H_2S was detected in the plume, which is consistent with thermodynamic calculations showing that the gas phase should have $SO_2/H_2S>10^4$ if buffered by the fO_2 of the magma [1, 2]. The compositions of the gas and scoria suggest a $\Delta^{34}S_{melt-gas}$ of +2.9±0.7‰ at T=1130°C [4]. This value is much larger than would be expected from experimentally-based gas-melt fractionation models (e.g.[2]), which would predict a $\Delta^{34}S_{melt-gas}$ of +0.5‰ at magmatic T and fO_2 [2, 4]. Degassing modeling using commonly employed fractionation factors [2] assuming 75% S loss from the melt [5] at 1100°C and fO_2 of ΔQFM +2 [2, 4] would predict an initial isotope composition of the magma of +7.2‰. However, if the observed $\Delta^{34}S_{melt-gas}$ value is used, the undegassed composition would be +3.8‰, implying far less contamination of the mantle source by subducted seawater sulfate. Analysis of S concentration and speciation in MA glasses will further constrain degassing fractionation.

The $\delta^{34}S$ of sulfate formed in and deposited from the SO_2 plume decreases exponentially with distance from the crater, from +7.5‰ at the crater to +4.9‰ at 14 km, indicating Rayleigh distillation of sulfate. Using the observed $\Delta^{34}S_{gas-aerosol}$ of +2.4‰ and wind speed of 5.9m/s, the rate of SO_2 to sulfate conversion is calculated at ~54%/hour. Distally measured SO_2 fluxes may therefore be underestimated.

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