Global CO_2 emission from volcanic lakes

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ABSTRACT

The global CO_2 discharge from subaerial volcanism has been estimated at ~300 Mt yr^{-1}. However, estimates of CO_2 emissions from volcanic lakes have not been considered. In order to improve this information, extensive research on CO_2 emissions of volcanic lakes worldwide has been performed. The observed normalized average CO_2 emission rates increase from alkaline (5.5 t km^{-2} d^{-1}), to neutral (201.2 t km^{-2} d^{-1}), to acid (614.2 t km^{-2} d^{-1}) in volcanic lakes. Taking into account (1) normalized CO_2 emission rates, (2) the number of volcanic lakes in the world (~769), and (3) the fraction and average areas of the investigated alkaline, neutral, and acid volcanic lakes, the estimated global CO_2 emission from volcanic lakes is 117 ± 19 Mt yr^{-1}, with 94 ± 17 Mt yr^{-1} as deep-seated CO_2. This study highlights the importance of a revision of the actual global CO_2 discharge from subaerial volcanism.

INTRODUCTION

During the past two decades, scientists have paid more attention to the volcanic emissions of CO_2 and its contribution to the global C budget (Gerlach and Graeber, 1985; Gerlach, 1991a, 1991b; Le Cloarec and Marty, 1991; Varekamp et al., 1992; Allard, 1992; Williams et al., 1992; Sano and Williams, 1996; Marty and Tolstikhin, 1998; Kerrick, 2001; Mörner and Etíope, 2002). Excluding mid-ocean ridges as a net source of CO_2 to the atmosphere, the total CO_2 discharge from subaerial volcanism has been estimated at 300 Mt yr^{-1} (Mörner and Etíope, 2002), accounting for both visible emanations and diffuse emissions from the flanks of volcanoes. Because this emission rate is lower than estimates of the global consumption of atmospheric CO_2 by subaerial silicate weathering, a revision of the estimates of CO_2 discharges from subaerial volcanism is necessary (Varekamp and Thomas, 1998; Kerrick, 2001). Other CO_2 sources might also balance the global carbon cycle, such as nonvolcanic CO_2 degassing (Chiodini et al., 2010) and oxidation of methane emitted from serpentinization of ultramafics (Kerrick, 2001), contributing in this way to the additional CO_2 necessary to balance the global carbon cycle. However, among all these studies, CO_2 emissions from volcanic lakes have never been taken into account in the global C budget balance from the solid Earth.

Volcanic lakes are generally formed by one of three mechanisms: (1) explosive excavation (crater lakes), (2) collapse (caldera lakes), and (3) blockage of common waterways (rivers, streams) by mudflows, lava flows, or ash (see *Overview of Volcanic Lakes*: http://www.wesleyan.edu/ees/JCV/vloverview.html). After the Monoun (1984) and Nyos (1986) volcanic lake gas disasters, both in Cameroon (Sigurdsson et al., 1987; Sigvaldason, 1989), the accumulation of CO_2 in volcanic lakes is a process well known in the scientific community, which realized the potential geological hazard that volcanic lakes represent (Le Guern and Sigvaldason, 1989, 1990; Evans et al., 1994; Kling et al., 2005; Kusakabe et al., 2008). Because large amounts of magmatic gases are dissolved in the water, CO_2 degassing from volcanic lakes should be taken into account in the global C budget.

Estimates of global CO_2 emission from volcanic lakes imply the need for a revision of the number of volcanic lakes on Earth for the following reasons: (1) the numbers listed in the literature (Delmelle and Bernard, 2000) differ significantly from the real numbers in several volcanic regions (e.g., there are ~22 volcanic lakes in the Azores, but only three of these were listed by Delmelle and Bernard [2006]; see Cruz et al. [2006]); (2) the lack of volcanic lakes in other volcanic regions listed by Delmelle and Bernard (2006) (e.g., the East African Rift Zone); and (3) the fact that many volcanoes host more than one volcanic lake (i.e., Rotorua in New Zealand). Therefore, a more complete revision of the potential number of volcanic lakes in the world has been done in this study.

METHODS

In order to evaluate the global CO_2 emission from volcanic lakes, an extensive survey of CO_2 emission at the surface environment of volcanic lakes was carried out between 2006 and 2010. For this study 31 observations from 24 volcanic lakes located in Nicaragua, Guatemala, El Salvador, Costa Rica, Japan, Cameroon, Philippines, France, and Germany were performed (Fig. 1). The volcanic lakes were chosen randomly following a simple random sampling in order to have a statistically representative data set. Other published data from volcanic lakes were also considered in this study (Table 1). Selected volcanic lakes were grouped, following the classification described by Pasternack and Varekamp (1997), into acid, neutral, and alkaline lakes; this approach was used in order to classify the studied lakes on the basis of their activity.

Since gas emissions through the lake surface occur by convective or advective degassing and/or by diffusion through the water-air interface (Mazot and Taran, 2009), CO_2 efflux measurements from volcanic lakes were performed using a floating device with an accumulation chamber and an infrared sensor with an accuracy of ~5% (Fig. 2). The reproducibility for the range 100–10,000 g m^{-2} d^{-1} is 10%. This random error is based on the uncertainty calculated from the variability of the measurements carried out in the laboratory. In order to convert volumetric concentrations to mass concentrations (g m^{-2} d^{-1}), atmospheric pressure, temperature, and total volume were taken into account. The field work was performed under dry and stable meteorological conditions. In each survey, a homogeneous sampling site distribution was designed; spacing between sites depended on the lake dimensions. At each observation point, water temperature and pH were measured at 20–40 cm depth by means of a thermocouple and a portable pH meter.

RESULTS AND DISCUSSION

To quantify the CO_2 emission from each volcanic lake, CO_2 efflux maps were constructed using conditional sequential Gaussian simulations provided by the GSLIB (Geostatistical Software Library) program (Deutsch and Journel, 1998; Cardellini et al., 2003). We performed ~200 simulations for each survey following the variogram model. An average map was then constructed for each survey using the average of the different values simulated at each cell. Because quantification of the uncertainty of the CO_2 emission is important, the mean and the

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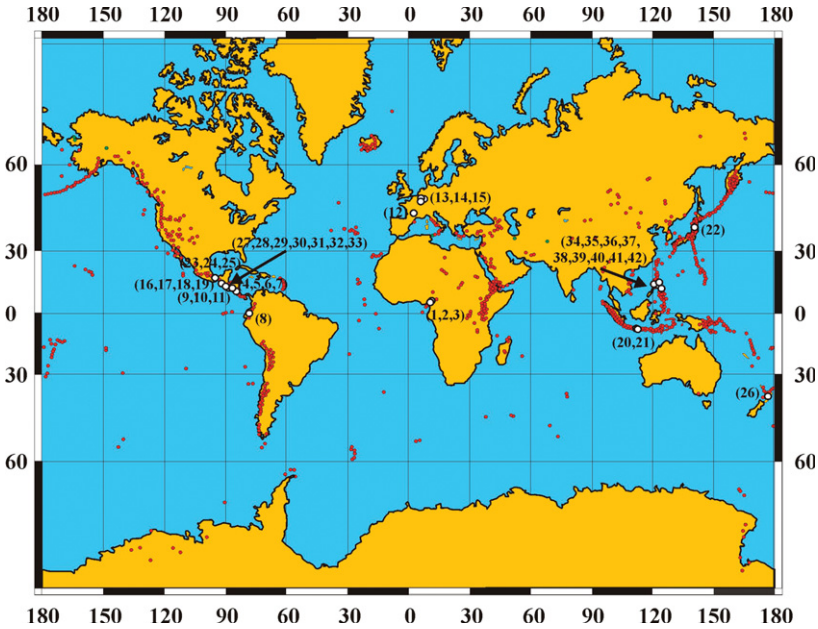

Figure 1. Locations of studied volcanic lakes (see Table 1). Red circles indicate Holocene volcanoes and white filled circles indicate studied volcanic lakes.

Figure 2. In situ CO_2 efflux measurements at surface environment of volcanic lakes with modified accumulation chamber method.

TABLE 1. LIST OF VOLCANIC LAKES CONSIDERED IN THIS STUDY

No.	Volcanic lake	Year	Location	CO_2 ($t\ d^{-1}$)		Area (km^2)	CO_2 ($t\ km^{-2}\ d^{-1}$)	Type	Pop.
1	Nyos[1]	2002	Cameroon	12.05	n. d.	1.80	6.70	alkaline	Pop. II
2	Nyos*	2009	Cameroon	16.69	±0.1	1.64	10.18	alkaline	Pop. II
3	Monoun*	2009	Cameroon	19.13	±0.1	0.63	30.37	neutral	Pop. III
4	Laguna de Botos*	2006	Costa Rica	0.12	±0.01	0.11	1.12	neutral	Pop. I
5	Laguna de Botos*	2010	Costa Rica	0.85	±0.46	0.11	7.73	acid	Pop. II
6	Laguna Río Cuarto*	2010	Costa Rica	31.40	±0.3	0.46	68.26	neutral	Pop. III
7	Laguna de Hule*	2010	Costa Rica	100.00	±1.8	0.55	182.82	neutral	Pop. III
8	Cuicocha[2]	2006	Ecuador	106.00	±5	13.25	8.00	alkaline	Pop. II
9	Ilopango[3]	1999	El Salvador	1111.00	n. d.	99.50	11.17	alkaline	Pop. II
10	Crater Santa Ana[4]	2002	El Salvador	7.00	n. d.	0.03	222.93	acid	Pop. III
11	Coatepeque*	2006	El Salvador	375.00	n. d.	24.83	15.10	neutral	Pop. III
12	Lac Pavin*	2007	France	0.01	±0.003	0.47	0.02	alkaline	Pop. I
13	Shalkermehren*	2007	Germany	0.11	±0.01	0.25	0.43	alkaline	Pop. I
14	Germundeer*	2007	Germany	0.32	±0.01	0.10	3.16	alkaline	Pop. I
15	Laacher See*	2007	Germany	35.10	±1.7	2.50	14.04	neutral	Pop. III
16	Laguna de caldera*	2008	Guatemala	0.01	±0.001	0.40	0.03	alkaline	Pop. I
17	Atitlan*	2008	Guatemala	416.00	±4.7	119.90	3.47	alkaline	Pop. II
18	Amatitlan*	2008	Guatemala	67.00	±17.6	13.87	4.86	alkaline	Pop. II
19	Ixpaco*	2008	Guatemala	85.60	±14.8	0.10	856.00	acid	Pop. IV
20	Kelud[5]	2001	Indonesia	105.00	n. d.	0.11	1000.00	neutral	Pop. IV
21	Kelud[6]	2006	Indonesia	35.00	n. d.	0.11	333.33	neutral	Pop. III
22	Katanuma crater lake*	2010	Japan	17.00	±0.6	0.14	121.43	acid	Pop. III
23	El Chichon[6]	2007	Mexico	164.00	±9.5	0.14	1188.41	acid	Pop. IV
24	El Chichon[7]	2007	Mexico	59.00	±2.5	0.08	724.82	acid	Pop. IV
25	El Chichon[7]	2008	Mexico	144.00	±5.9	0.10	1484.54	acid	Pop. IV
26	Ruapehu[8]	2010	New Zealand	92.00	n. d.	0.21	438.10	acid	Pop. III
27	Apoyo*	2006	Nicaragua	539.00	±6	21.00	25.67	alkaline	Pop. III
28	Tiscapa*	2006	Nicaragua	4.00	±0.24	0.14	28.57	neutral	Pop. III
29	Jiloa*	2006	Nicaragua	734.00	±14	3.75	195.73	neutral	Pop. III
30	Laguna de Masaya*	2009	Nicaragua	868.90	±15.8	8.30	104.69	neutral	Pop. III
31	Asososca Managua*	2009	Nicaragua	7.25	±0.12	0.77	9.42	neutral	Pop. II
32	Apoyeque *	2010	Nicaragua	211.70	±12.9	2.60	81.42	neutral	Pop. III
33	Nejapa*	2010	Nicaragua	5.80	±0.54	0.2	29.00	neutral	Pop. III
34	Taal caldera*	2008	Philippines	823.00	±14	227.00	3.63	alkaline	Pop. II
35	Pinatubo crater lake*	2008	Philippines	711.00	±16.8	2.00	355.50	neutral	Pop. III
36	Taal crater lake*	2008	Philippines	506.00	±14.9	1.29	392.25	acid	Pop. III
37	Laguna de Danao*	2009	Philippines	0.03	±0.01	0.12	0.28	alkaline	Pop. I
38	Pinatubo crater lake*	2009	Philippines	928.00	±18.9	2.00	464.00	neutral	Pop. III
39	Taal crater lake*	2009	Philippines	948.00	±21.9	1.29	734.88	acid	Pop. IV
40	Lago de Buhi *	2009	Philippines	0.41	±0.3	16.90	0.02	alkaline	Pop. I
41	Pinatubo crater lake*	2010	Philippines	1014.00	±16	2.00	507.00	neutral	Pop. IV
42	Taal crater lake*	2010	Philippines	763.00	±18	1.29	591.47	acid	Pop. IV

Note: Pop.—population; n.d.—no data available. Data sources: *This study; 1—Kusakabe et al. (2008), 2—Padrón et al. (2008), 3—López et al. (2004), 4—Bernard et al. (2004), 5—Mazot (2005), 6—Mazot and Taran (2009), 7—Mazot et al. (2010), 8—A. Mazot (2010, personal commun.).

standard deviations of the 200 simulated values of total CO_2 output were assumed to be the characteristic values of the CO_2 released and its uncertainty (Cardellini et al., 2003). Figure 3 shows the CO_2 efflux distribution map of the survey carried out at Pinatubo volcanic lake in 2008 based on the mean simulated total CO_2 output value. Table 1 shows the calculated mean of the CO_2 output values for each survey.

To estimate the global CO_2 emission from volcanic lakes, two approaches were applied after normalizing the CO_2 emission rates from each volcanic lake by the area. For the first approach volcanic lakes were grouped following the classification of acid, alkaline, and neutral lakes, and the arithmetic mean of the CO_2 emission rate for each group was calculated. The observed highest emission rate was for acid (614.2 t km^{-2} d^{-1}), followed by neutral (201.2 t km^{-2} d^{-1}), and then alkaline volcanic lakes (5.5 t km^{-2} d^{-1}). Alkaline volcanic lakes showed the lowest emission rate; almost all the CO_2 is present as HCO_3^- and CO_3^{2-} in the water, and the fraction of dissolved CO_2 is generally very small. So, although the alkaline volcanic lakes may be carbonate rich, only a small fraction escapes through the surface to the atmosphere, and the rest may leave the lake system through carbonate precipitation or seepage. Even when CO_2 emissions at individual acid volcanic lakes are higher than at neutral and alkaline ones, the total CO_2 contribution of each group depends mainly on the number of lake types and their area, which is demonstrated in this study (part A in Table 2). If we consider the arithmetic mean values of the areas for each group as the representative area for each group of lake type, the contribution of CO_2 emission rate for each group can be calculated by multiplying by the total number of volcanic lakes in the world, ~769. This potential number of volcanic lakes on Earth is the result of multiplying the number of listed volcanic lakes in the literature (138; Delmelle and Bernard, 2000), times the average ratio of the number of observed volcanic lakes to the number of listed volcanic lakes in the literature (ratio = 5.6) from 17 different volcanic regions worldwide. Assuming that 32 volcanic lakes of this study could be a good representative data set of the world volcanic lakes, we calculated the CO_2 emission rate fraction of each group. Summing the CO_2 emission rates from the three groups, the global CO_2 emission from volcanic lakes gives a total output of 104 Mt yr^{-1} (part A in Table 2), ignoring all the uncertainties and time variations of individual flux data.

The second approach to estimate the global CO_2 emission from volcanic lakes was basically to evaluate the existence of different populations in the normalized CO_2 emission data by means of a statistical-graphical analysis (Sinclair, 1974). This approach provides a fundamental grouping of data values and can be applied to any polymodal distribution, allowing us to extract individual populations from a polymodal distribution. The distribution of normalized CO_2 emissions of volcanic lakes differs from a log-normal distribution, indicating that there is more than one population. The log probability plot of normalized CO_2 emissions of volcanic lakes indicates the existence of four different populations (Fig. 4). Following this grouping and multiplying the average normalized CO_2 emission rates for each population, times the arithmetic mean values of the areas for each population, times the cumulative percentage of each population, times the number of volcanic lakes in the world (~769), it has been estimated that the global CO_2 emission from volcanic lakes is 131 Mt yr^{-1} (part B in Table 2).

Therefore, we have considered the average value of both estimates, 117 ± 19 Mt yr^{-1} (range from 104 to 131 Mt yr^{-1}), as the global CO_2 emission from volcanic lakes. Estimates of the deep-seated CO_2 fraction of this global CO_2 emission from volcanic lakes could be

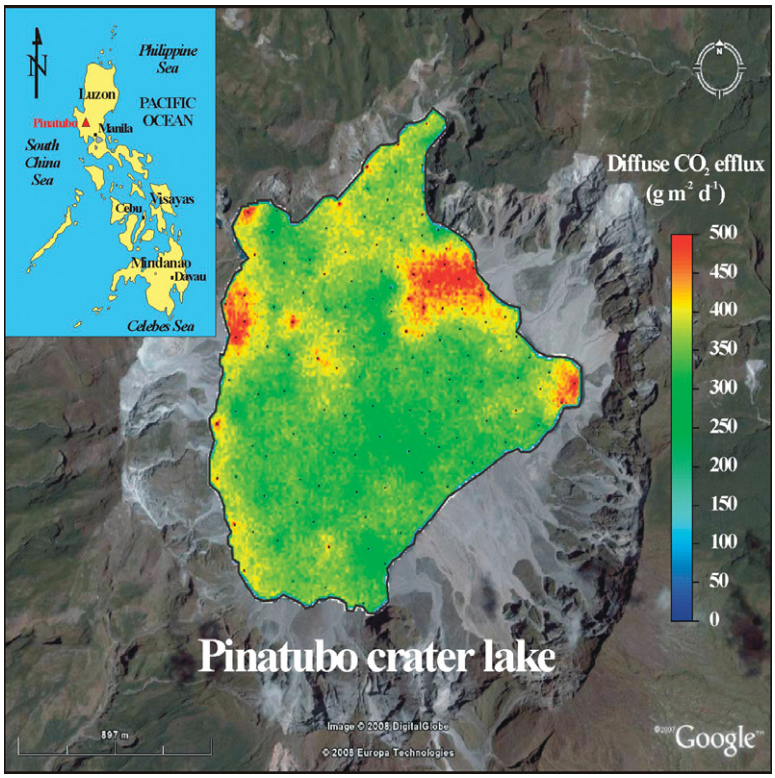

Figure 3. Spatial distribution of CO_2 emission from Pinatubo volcanic lake, Philippines.

TABLE 2. ESTIMATED GLOBAL CO_2 EMISSIONS BASED ON TWO DIFFERENT APPROACHES

A Volcanic lakes	Average CO_2 (t km^{-2} d^{-1})	Type (%)	Average area (km^2)	CO_2 emission (Mt yr^{-1})	CO_2 emission (%)
Acid	614.2	24	0.24	10	9.9
Neutral	201.2	38	3.35	72	69.2
Alkaline	5.5	38	36.87	22	20.9
Estimated global CO_2 emission from volcanic lakes				104	100.0
Estimated deep-seated CO_2				82	79.0
Estimated non deep-seated CO_2				23	21.0
B Volcanic lakes	**Average CO_2 (t km^{-2} d^{-1})**	**Type (%)**	**Average area (km^2)**	**CO_2 emission (Mt yr^{-1})**	**CO_2 emission (%)**
Pop. IV	884.8	25	0.64	40	30.4
Pop. III	172.4	35	3.93	66	50.4
Pop. II	7.2	23	53.09	25	19.1
Pop. I	0.7	17	2.62	0.1	0.1
Estimated global CO_2 emission from volcanic lakes				131	100.0
Estimated deep-seated CO_2				106	80.8
Estimated non-deep-seated CO_2				25	19.2

Note: A: Normalized CO_2 emission rates from acid, neutral, and alkaline type; B: four different observed populations (Pop.) in volcanic lakes.

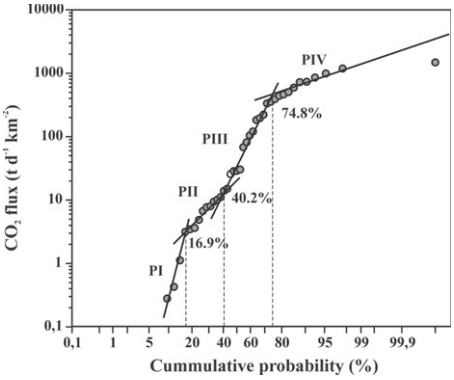

Figure 4. Probability plot of normalized CO_2 emission rates from studied volcanic lakes. P—population.

mainly associated to the contributions from acid and neutral volcanic lakes (82 Mt yr^{-1}) as well as from populations III and IV (106 Mt yr^{-1}) for the first and second approach, respectively (Table 2). Therefore, an average of 94 ± 17 Mt yr^{-1} could be considered an estimate of the global deep-seated CO_2 emission from volcanic lakes, and it does not discriminate between the fraction of nonvolcanic CO_2 fed by local ground and surface waters and the mixing ratios of volcanic fluids and meteoric fluids. This value is close to 30% of the estimate for the global CO_2 emission from subaerial volcanoes, 300 Mt yr^{-1} (Mörner and Etíope, 2002), and highlights the importance of a revision of the total CO_2 discharge from subaerial volcanism, which seems to be underestimated and should be taken as a very conservative lower limit.

Our results show that the global deep-seated CO_2 emission from volcanic lakes is similar to the estimated for submarine volcanism (mid-ocean ridges), 66–97 Mt yr^{-1} (Varekamp et al., 1992; Marty and Tolstikhin, 1998), and six times lower than the estimated value of lithospheric degassing of CO_2 that is added directly to the atmosphere at a rate of 600 Mt yr^{-1} (Mörner and Etíope, 2002).

Assuming that the value of 117 ± 19 Mt yr^{-1} is representative of global CO_2 emission from volcanic lakes, we would suppose that no more than 0.32% of the actual CO_2 emission is from anthropogenic sources, ~36,300 Mt yr^{-1} (Le Quéré et al., 2009); despite that, this amount should be taken into account for a refined global carbon budget modeling.

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