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CO<sub>2</sub> ADSORPTION ON VOLCANIC ASH: NET CONTRIBUTION OF VOLCANIC  
ERUPTIONS TO THE GLOBAL CARBON CYCLE

A Thesis  
presented in partial fulfillment of requirements  
for the degree of Master of Science  
in the department of Geology and Geological Engineering  
The University of Mississippi

by

LEE NEW

August 2017

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## ABSTRACT

Adsorption of CO<sub>2</sub> on mineral surfaces accounts for a small percentage of the terrestrial CO<sub>2</sub> sink. Volcanic ash is virtually free of CO<sub>2</sub> when erupted and has a high surface area resulting in a high capacity for CO<sub>2</sub> adsorption. Volcanism is commonly assumed to be only a source of CO<sub>2</sub>, but pyroclastic eruptions can also potentially serve as a sink.

Adsorption was investigated for 6 volcanic ash samples of andesitic composition from four volcanoes: Okmok, Redoubt, Spurr, and Tungurahua. The eruption of Volcano Redoubt released  $0.75 \times 10^9$  metric tons of CO<sub>2</sub> and  $55 \times 10^9$  kg of ash throughout the 3 week eruption. The eruption of Mount Spurr released 85,000 CO<sub>2</sub> and  $36 \times 10^9$  kg of ash throughout the duration of the eruption. The eruption of Volcano Tungurahua released  $29 \times 10^9$  kg of ash. CO<sub>2</sub> emissions from the eruption of Volcano Tungurahua were not measured. CO<sub>2</sub> emissions and mass of tephra released from the eruption of Mount Okmok were not measured. The calculations for adsorption percent for these two volcanos were performed using the ash/CO<sub>2</sub> ratio from Mount Spurr and Volcano Redoubt. The ratio was then applied to Okmok and Tungurahua to solve for CO<sub>2</sub> emissions for each of these ratios. The calculations for CO<sub>2</sub> adsorption for Mount Okmok were performed using the mass of tephra from Volcano Kasatochi (an adjacent, similar erupting volcano). Adsorption on each of the six ash samples ranged from 34.3 to 67.2 mg of CO<sub>2</sub> per kg of ash. Adsorption ranged from 0.04 to 0.45 mg CO<sub>2</sub> per m<sup>2</sup> volcanic ash.

## ACKNOWLEDGEMENTS

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## I. INTRODUCTION

Modeling global climate change depends on an accurate knowledge of carbon sources and sinks. Major sources of CO<sub>2</sub> to the atmosphere include volcanic eruptions, respiration by living organisms, decay of organic material, deforestation, cultivation of land, and anthropogenic activity, including land use changes, fossil fuel combustion, and cement production (Gerlach, 2011, Denman et al., 2007, Achard et al., 2004, Houghton, 2003, Victor et al., 2014, Pachauri, 2014). Possible carbon sinks investigated have included forest biomass (e.g. Hopkinson et al., 2016; Luysaert et al., 2008; McMahon et al., 2010), weathering reactions (Brady, 1991; Andrews and Schlesinger, 2001; Dessert et al., 2003; Liu et al., 2011), soil organic material (Gaudinski et al., 2000; Harrison, 2004; Brown et al., 2010), volcanic soils (Zehetner, 2010), erosion and burial of carbon (Stallard, 1998; Einsele et al., 2001; Downing et al., 2008), soil pore water (Andrews and Schlesinger, 2001), adsorption on mineral surfaces (Davidson et al., 2013), and even bacteria (Braissant et al., 2002). There is currently more CO<sub>2</sub> removed from the atmosphere than accounted for by known sinks. Denman et al. (2007) estimate the size of the residual terrestrial sink (formally known as the missing sink) to be 2-3 Pg C yr<sup>-1</sup>.

CO<sub>2</sub> is the second most abundantly emitted volcanic gas behind water vapor (Casey, 2010). Estimates of CO<sub>2</sub> emissions from divergent plate, intraplate, and convergent plate subaerial and submarine volcanoes range between 0.15 and 0.26 Gt/y (Gerlach, 2011). More specific emission data is available for a few specific volcanic events. The 1991 eruption of Mount Pinatubo added an estimated 0.0005 Gt of CO<sub>2</sub> to the atmosphere. The 1980 eruption of Mount St. Helens contributed an estimated 0.01 Gt (Gerlach, 2011). Long term continuous degassing also

contributes to volcanic CO<sub>2</sub> emissions. The central Oregon Cascades are degassing at an estimated emission rate of 0.0000113 Gt/yr. There is a lack of data estimating volcanic CO<sub>2</sub> emissions. The emissions that have been reported are based on statistical estimates projected from isolated measurements. Volcanic systems are complex and highly variable, creating significant uncertainty in these estimates. Volcanism is often assumed to be only a source of CO<sub>2</sub>, but pyroclastic eruptions can also serve as a sink.

Previous studies have shown evidence of CO<sub>2</sub> adsorption on various substrates including basalts, tuffs, and other sediments (e.g., Zent et al., 1987; Streigl and Armstrong, 1990; Davidson et al., 1995). Davidson et al. (2013) quantified the portion of the carbon sink for North America due to adsorption on soils and volcanic rock in response to rising atmospheric CO<sub>2</sub> concentrations. Volcanic ash in this study proved to have a considerably higher adsorption capacity than samples of other volcanic substrates, leading to speculation that a significant portion of CO<sub>2</sub> released during an eruption could be reabsorbed. The volcanic ash erupted from a volcano will be virtually free of CO<sub>2</sub> at high temperature, and have an extremely high adsorption capacity as it cools. The results from this study may address the question: What is the net addition of volcanic CO<sub>2</sub> after accounting for subsequent adsorption on freshly deposited ash?

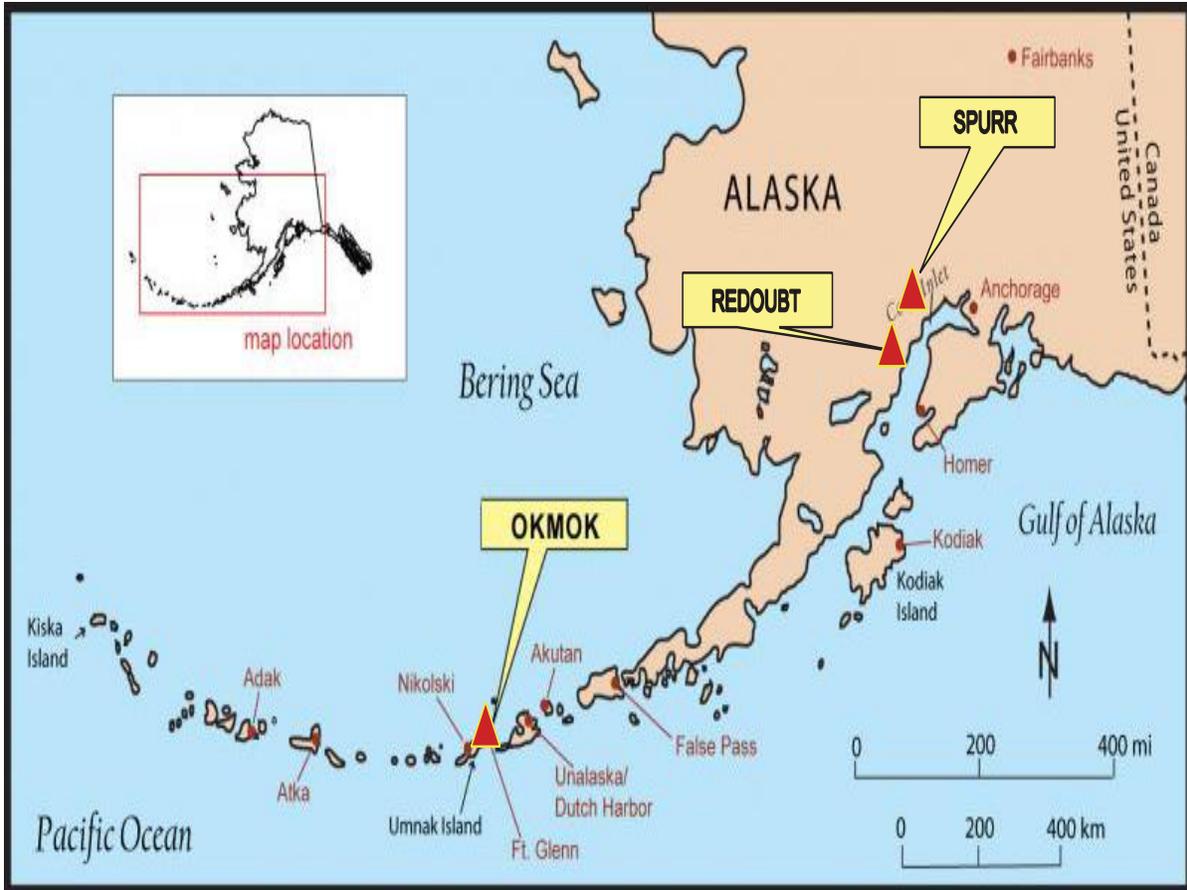
The current study quantified CO<sub>2</sub> adsorption on six volcanic ash samples collected from four eruptions. Duplicate samples collected from different locations for the same eruption give an idea of variability between sampling locations. All samples were predominantly andesitic in composition, which is not due to biased sampling. Mafic and basaltic samples were not specifically excluded. Volcanic eruptions that produce basaltic rocks are typically nonexplosive. These eruptions have relatively low silica contents and rarely generate large volumes of ash.

Explosive eruptions that create ash tend to result from volcanos with higher silica contents and more viscous magmas. This type of eruption produces ash of andesitic, dacitic, and rhyolitic composition. Due to availability of preserved ash samples, volcanic ash of andesitic composition were the only samples acquired. Carbon dioxide adsorption at atmospheric CO<sub>2</sub> concentrations was quantified for each sample measured at 420 ppm. This is slightly higher than reported CO<sub>2</sub> concentrations. The mass of measured adsorbed carbon for each ash sample was used with estimates of emitted CO<sub>2</sub> and ejected tephra to calculate the potential net removal by volcanic ash. The goal is to determine what fraction of CO<sub>2</sub> is adsorbed.

## II. GEOLOGIC SETTING AND MINERALOGY

### SAMPLE SELECTION

CO<sub>2</sub> adsorption experiments were conducted on six samples of volcanic ash from four different eruptions. The locations are identified in Figures 1 and 2. Ash samples were collected by geologists in the field near the time of eruption and stored in air tight containers. The eruption date and duration, collection date, and sample location are displayed in Table 1. Sample RDASH1 was collected during the 2009 eruption of Redoubt Volcano. This sample was recovered from fallout on April, 4<sup>th</sup> 2009, the date of the eruption, 75 km SE the vent in Homer, Alaska. Sample MSASH1 was collected from the 1992 eruption of Mount Spurr. MSASH1 was recovered on August, 18<sup>th</sup> 1992 in Anchorage, Alaska, from fallout 120 W km from Crater Peak vent. Samples OKASH1 and OKASH2 were collected from the 2008 eruption of Mount Okmok. Both samples were recovered after fallout in September, 2008, after a five-week eruption on Umnak Island, one in Fort Glenn, Alaska, 10 km SE of the vent (OKASH1), and 4 km SE of the vent near Fort Glenn (OKASH 2). Samples TASH1 and TASH2 were collected from the 2014 eruptions of Tungurahua in the Cordillera Oriental of Ecuador. The researchers recovered TASH1 on February 1<sup>st</sup>, 2014, on the day of the eruption in Palitagua, Ecuador, 6 km SW of the vent. TASH2 was recovered from the Mayou station on the SE slope 4 km from the vent on April 8<sup>th</sup>, 2014, three days after the eruption. The following descriptions and properties of each ash sample are summarized in Table 2.



**Figure 1. Location of Alaskan Volcanoes**



Figure 2. Location of Ecuadorian Volcano

**Table 1. Volcano eruption and collection data (see text for references).**

	<b>Eruption Date</b>	<b>Eruption Duration</b>	<b>Collection Date</b>	<b>Sample Location</b>
<b>Redoubt, AK</b>	April 4, 2009	3 Wks	April 4, 2009	75 Km from vent - Homer
<b>Spurr, AK</b>	August 18, 1992	4 Hrs	August, 1992	120 Km from vent - Anchorage
<b>Okmok, AK</b>	July 12, 2008	5 Wks	September, 2008	10 Km SE of crater - Fort Glen
				4 Km SE of vent
<b>Tungurahua, Ecuador</b>	February 1, 2014	cycles of intermitted eruptions	February 1, 2014	6 Km SW of vent - Palitugua
	April 5, 2014		April 8, 2014	4 Km SE of vent - Mayou

**Table 2. Volcanic ejecta data (see text for references).**

	<b>Plume Height (Km)</b>	<b>Tephra Mass (Kg)</b>	<b>Composition</b>	<b>Silica Content (%)</b>	<b>Grain Size (mm)</b>
<b>Redoubt, AK</b>	19	$55 \times 10^9$	Andesite	59-62	0.125
<b>Spurr, AK</b>	14	$36 \times 10^9$	Andesite	62	0.5 - 0.7
<b>Okmok, AK</b>	16	$600 \times 10^9$	Basaltic Andesite	55-57	2.0 - 3.0
<b>Tungurahua, Ecuador</b>	15	$29 \times 10^9$	Andesite	58-59	0.375

## ERUPTION DATA AND MINERALOGY

### *Volcano Redoubt, Homer, Alaska: RDASHI*

Redoubt Volcano erupted for over two weeks resulting in 20 tephra-producing explosions. The April 4<sup>th</sup> 2009 eruption, from which RDASH was collected, resulted in substantial tephra fall. The subplinian eruptions resulted in plume heights up to 19 km above sea level and tephra deposits totaling a volume of  $21 \times 10^6 \text{ km}^3$  and a mass of  $55 \times 10^9 \text{ kg}$ . The wind-carried plume traveled as far as Fairbanks (Wallace et al., 2013). The eruption of Volcano Redoubt released 20,000 t/d  $\text{CO}_2$  on the day of the eruption and released  $0.75 \times 10^6$  metric tons during the eruption duration.  $\text{CO}_2$  data was collected by an unpressurized piper PA-31 Navajo fixed wing aircraft with a LICOR LI-7000 infrared  $\text{CO}_2$  analyzer (Werner et al., 2013). The significant volume of tephra was comprised of aggregates, fine ash clusters (0.125 mm) and consists mostly of medium-gray, low-silica andesitic scoria. The scoria made up 45–50% of sampled ash deposits (Wallace et al., 2013).

### *Crater Peak Vent, Mount Spurr, Anchorage, Alaska: MSASHI*

The August 18<sup>th</sup>, 1992 eruption of Mount Spurr was the last in a series of three. The subplinian eruption lasted 4 hrs and mainly consisted of tephra fall, along with less significant pyroclastic flows, debris flows, and ballistic showers (Harbin et al., 1992). This single eruption resulted in a tephra fall volume of  $56 \times 10^6 \text{ km}^3$  (Neal et al., 1994) and a mass of  $36 \times 10^9 \text{ kg}$  (McGimsey, 2001). The Aug 18 eruption was the most violent, propelling ash 14 km into the air where it was carried to Anchorage and the Yukon Territory of Canada over 860 km from the peak (Neal et al., 1994; Swanson et al., 1992).  $\text{CO}_2$  monitoring of the eruption of Mount Spurr began on September, 25<sup>th</sup> five weeks after the eruption.  $\text{CO}_2$  concentrations were measured at

11,000 metric tons the first day of measurement. The concentrations continued to slowly decrease to background levels for the next two weeks. Over this 12 day monitoring period, Mount Spurr released an estimated 85,000 metric tons of CO<sub>2</sub>. CO<sub>2</sub> data was collected by a fixed wing aircraft with an airborne infrared spectrometer measurement (MIRAN) and airborne ultraviolet correlation spectrometer (CO-SPEC) (Doukas and Gerlach, 1995). The ash was primarily described as high-silica andesite. Silica dominated the composition (62% by mass) (Harbin et al., 1992; Swanson et al., 1992). The grain size ranged in diameter from 0.5 to 0.7 mm, and the color ranged from light gray to brown (Swanson et al., 1992). The porphyritic andesite occurred in blocky fragments with a microlite matrix with embedded phenocrysts of plagioclase, hornblende, augite, hypersthene, and FeTi oxides (Swanson et al., 1992). The remainder of ash is composed of high-silica (69–73% silica), light brown or colorless dacitic and rhyolitic glass (Swanson et al., 1992).

*Okmok Caldera, Umnak Island, Alaska: OKASH1 and OKASH2*

The eruption of the Okmok volcano was the first of two in the Aleutian Islands during the summer of 2008. The subplinian Okmok eruption was the largest. Okmok erupted for five weeks resulting in an initial plume height of 16 km, and a tephra fall volume totaling  $400 \times 10^6$  km<sup>3</sup> (Langmann et al., 2010; Larsen et al., 2015). CO<sub>2</sub> emissions and mass of tephra released from the eruption of Mount Okmok were not measured. The eruptions resulted in scoria-rich fall deposits classified as basaltic andesite. The basaltic andesite ash consisted of 55–57% wt. silica. During the time of collection, the scoria ranged in size from 1–2 cm in diameter and was coated by very-fine-grain ash ranging from 2.0–3.0 mm in diameter (Larsen et al., 2015).

*Vulcano Tungurahua, Cordillera Oriental, Ecuador: TASH1 and TASH2*

Vulcano Tungurahua is one of the most active volcanoes in Ecuador. The strombolian eruptions during spring of 2014 were composed primarily of pyroclastic and tephra fall consisting mainly of andesitic ash. The majority of the tephra fall averaged 0.375 mm in diameter (Eychenne et al., 1992). The andesitic ash is 58–59% wt. silica, and the eruption produced an ash plume 15 km high. The eruption of brown andesitic ash resulted in a total mass of  $29 \times 10^9$  kg and reached distances of 200 km away from the vent (Bernard et al., 2016). The CO<sub>2</sub> emissions from the eruption of Volcano Tungurahua were not measured.

### III. METHODS

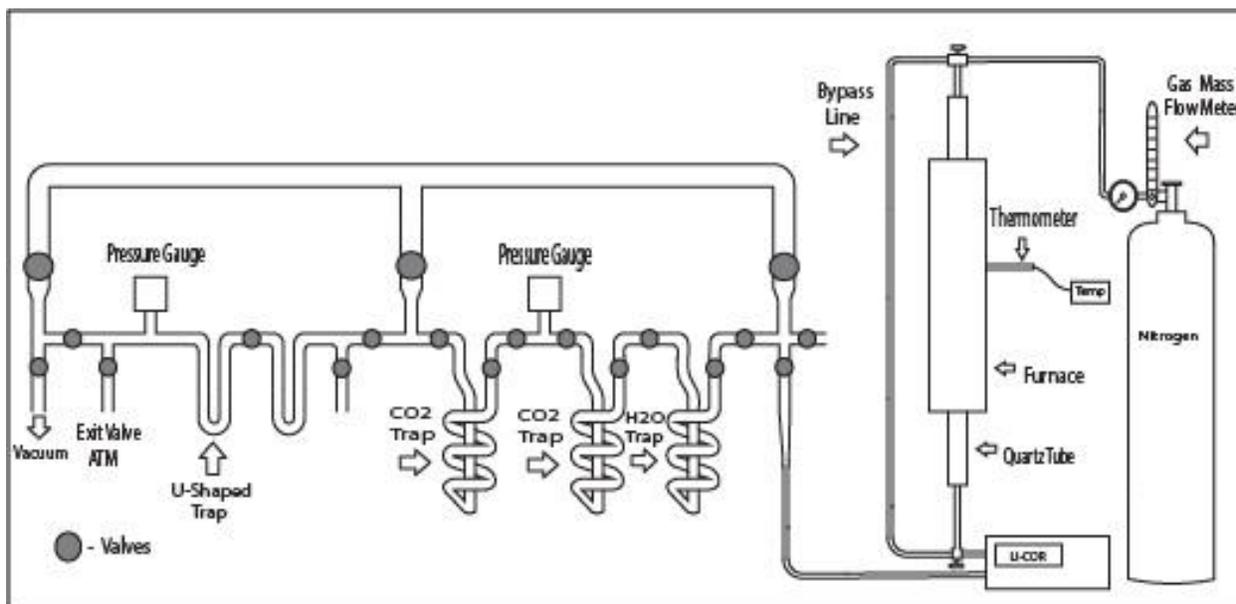
#### CHARACTERIZATION OF SURFACE AREA

Single point and multi-point BET analyses were performed on all 6 samples to determine the specific surface area of the volcanic ash. The samples were sent to Quantachrome's Material Characterization Laboratory. A BET (Brunauer, Emmett and Teller) surface area analysis with nitrogen was performed.

#### ADSORPTION APPARATUS

A fixed-bed adsorption system and glass vacuum line with cryogenic traps was assembled for all adsorption experiments (Figure 3). The adsorption system included a cylindrical quartz sample vessel and a ceramic furnace. The sample vessel was 0.6 m long with an outer diameter of 19 mm and an inner diameter of 13.5 mm. The 2.75 mm thick walls of the high-temperature quartz sample vessel allow the sample to be heated in the ceramic furnace to the temperature of 200° C. To keep the sample in place, quartz wool was placed above and below the ash sample. A series of valves connected the 6.35 mm outer-diameter copper tubing from a CO<sub>2</sub>-free nitrogen tank to the top of the column. When recording flow rates and concentrations were not necessary, the gas could be released to the atmosphere. When recording and collecting gases, the gas exiting the bottom of the column could be directed through a CO<sub>2</sub>/H<sub>2</sub>O analyzer (LICOR LI-7000). Next, the gas was streamed into the glass vacuum line for collection and out of the exit valve to the atmosphere. The LICOR was connected to a computer where software could log and record CO<sub>2</sub> and H<sub>2</sub>O concentrations, pressure, flow rate, and

temperature at various time intervals. The valves of the adsorption system could also direct gas to bypass the sample to flush the LICOR and vacuum line. The vacuum line was equipped with helical traps for H<sub>2</sub>O and CO<sub>2</sub> collection, valves for gas flow control, and two digital pressure sensors to measure collected mass.



**Figure 3. Fixed-bed adsorption system and glass helical trap vacuum apparatus**

## ADSORPTION

Atmospheric-CO<sub>2</sub> adsorption experiments were performed on each of the six ash samples. The ash sample was weighed and added to an Erlenmeyer flask. The flasks were placed in an oven for 24 hrs at 200°C to remove any preexisting CO<sub>2</sub>. Once the sample was baked, a rubber stopper was used to plug the flask. A peristaltic pump was used to pump atmospheric air with ambient CO<sub>2</sub> concentration and humidity through the rubber stopper into the flask and out a vent port. A filter with a 1 micron pore size was placed in the line to filter any debris and water. To establish an adequate equilibration time, each sample was exposed to atmospheric air for a variation of times ranging from 12 to 72 hours. By comparing the CO<sub>2</sub> concentrations of the sample after each equilibration time, an appropriate minimum time could be established. An increase in recovered carbon was observed when the equilibration time was increased from 12 to 24 hours; from 24 to 72 hours, recovered carbon did not increase. The equilibration time for subsequent samples was 24 hrs.

A similar process was performed to decide the size of the sample used for experimentation. The smallest sample size used (10 g) resulted in a low recovered mass of CO<sub>2</sub> too close to the system blank, introducing high uncertainty. The largest sample size used (40 g) resulted in a recovered mass of CO<sub>2</sub> that could over-pressurize the collection volume. The larger sample size also filled the majority of the sample vessel restricting the flow rate and increasing backpressure in the adsorption system. The recovered mass of CO<sub>2</sub> from a sample size of 20 g was determined to be the optimal size for this system.

The equilibrated sample was placed into the sample vessel, inserted vertically into the ceramic furnace, and connected to CO<sub>2</sub>-free nitrogen gas. The bypass line, LICOR, and vacuum line were alternately evacuated and purged with CO<sub>2</sub>-free nitrogen to remove residual CO<sub>2</sub> from

all parts of the system other than the sample vessel. Prior to desorption, helical trap 1 was immersed in dry ice and isopropyl alcohol to capture any desorbed water vapor; helical traps 2 and 3 were immersed in liquid nitrogen to capture desorbed CO<sub>2</sub>. The furnace was turned on, the sample vessel was opened to the traps, and CO<sub>2</sub>-free nitrogen was flushed at an estimated rate of 5-15 mL/min through the sample vessel, passing through the LICOR before the collection traps and discharging to the atmosphere. A tertiary trap in liquid nitrogen was placed at the exit point to ensure that atmospheric CO<sub>2</sub> did not migrate up-gradient into a sample trap. The CO<sub>2</sub> concentration, temperature, and flow rate was logged and recorded by the LICOR.

The desorption process was continued until the LICOR confirmed that the discharging CO<sub>2</sub> concentration reached zero. The desorption process typically took 6 hours. Once all of the CO<sub>2</sub> was captured, the vacuum line was sealed from helical trap two to the exit valve. Liquid nitrogen was placed around the U-shaped trap of the known-volume portion and removed from the traps. After allowing CO<sub>2</sub> to move from the helical to the U-shaped trap, the line was opened to the vacuum pump to remove any residual non-CO<sub>2</sub> gases. The known volume was sealed, and the liquid nitrogen was removed from the U-shaped trap. The final pressure change from the CO<sub>2</sub> sublimation inside the vacuum line was recorded and used in the calculation of the mass of CO<sub>2</sub>.

## BLANK EXPERIMENTS

Blank experiments were performed to measure CO<sub>2</sub> in the air space of the sample tube, and CO<sub>2</sub> adsorption on the sample tube walls or quartz wool. An empty sample vessel was packed with quartz wool. The vessel was then placed in the adsorption system and baked at 200°C for six hours (the typical bake off time). The same collection methods were performed with the blank experiments. The measured blank experiment was performed four times.

To determine if any CO<sub>2</sub> was diffusing in from the outside of the glass line or was not completely being flushed, liquid nitrogen was placed on the helical traps for six hours while CO<sub>2</sub>-free nitrogen was flushed through the line. This process was repeated, but no CO<sub>2</sub> was collected through diffusion or by improper flushing of the glass line.

A corrected blank was also calculated to account for the volume of the solids from the ash sample. The ash sample occupies a portion of the volume of the vessel resulting in an overestimate of CO<sub>2</sub> from the experimental blanks if uncorrected. The ash samples are of andesitic composition with a large portion consisting of silica. The specific gravity of andesitic rocks range is 2.5 to 2.8. The average (2.65) was used to calculate the volume of the solids in the sample vessel. The mass of CO<sub>2</sub> occupied by the volume of solids (M<sub>VS</sub>) was determined using:

$$M_{VS} = \left( \frac{P * V_S}{R * T} \right) (ATM_{CO_2})$$

Where M<sub>VS</sub> is moles of CO<sub>2</sub>, P is pressure (ATM), V<sub>S</sub> is the volume of the solids in the sample vessel (L), R is the gas constant ( $\frac{L*ATM}{mole*K}$ ), T is the temperature (K), and ATM<sub>CO<sub>2</sub></sub> is the atmospheric concentration of CO<sub>2</sub> (ppm). The final calculated blank (CB<sub>F</sub>) was determined using:

$$CB_F = MB_E - M_{VS}$$

Where CB<sub>F</sub> is moles of CO<sub>2</sub>, MB<sub>E</sub> is the experimental measured blank (moles), and M<sub>VS</sub> is mass of CO<sub>2</sub> from the volume of solids (moles). The final calculated blank was subtracted from the results of each adsorption experiment.

## ESTIMATES OF FRACTION OF CO<sub>2</sub> RELEASED

The mass of CO<sub>2</sub> (M<sub>CO2</sub>) adsorbed for each sample was determined using:

$$M_{CO_2} = \left( \frac{P * V}{R * T} \right) - CB_F$$

Where M<sub>CO2</sub> is moles, P is observed pressure change(ATM), V is the known volume in glass apparatus (L), R is the gas constant ( $\frac{L*ATM}{mole*K}$ ), T is the temperature (K), and CB<sub>F</sub> is the final calculated blank (moles). The mass of CO<sub>2</sub> adsorbed per kg of ash (M<sub>A</sub>) for each sample was determined using the following equation:

$$A = mol\ CO_2 * molecular\ weight\ of\ CO_2 * 1000\ mg$$

$$M_A = \frac{A}{B}$$

Where moles of CO<sub>2</sub> are converted to mg of CO<sub>2</sub>, M<sub>A</sub> is mass of CO<sub>2</sub> in mg per kg of ash, A is mg of CO<sub>2</sub>, and B is kg of ash used in each experiment. The adsorption capacity (AC) for a specific eruption was calculated using:

$$AC = AR \times M_A$$

Where AC is represented in metric tons, AR is mass of ash released from eruption (Kg), and M<sub>A</sub> is mg of CO<sub>2</sub> adsorbed per kg of ash. The adsorption percent (AP) was calculated using:

$$AP = \frac{AC}{GR}$$

Where AP is a percent, AC is adsorption capacity of the eruption (metric tons) and GR is mass of CO<sub>2</sub> released from the eruption (metric tons). If the mass of the tephra or mass CO<sub>2</sub> emitted

from an eruption is unknown, masses from a similar eruption were used to estimate adsorption capacity and percent of CO<sub>2</sub> emitted adsorbed.

## IV. RESULTS AND DISCUSSION

### SPECIFIC SURFACE AREA

Single point and multi-point BET analyses were performed on all 6 samples to determine the specific surface area of the volcanic ash (Table 3). The specific surface area of the samples ranged from 0.127 to 1.174 m<sup>2</sup>/g. The two samples from Mount Okmok were 0.539 and 0.548 m<sup>2</sup>/g. The two samples from Volcano Tungurahua were 0.127 and 0.148 m<sup>2</sup>/g. Given the variability of results from the four volcanos, the variability between two samples from the same volcano was small and fairly consistent.

### BLANK EXPERIMENTS

The experimental measured blanks were performed four times. The mass of CO<sub>2</sub> from the volume of solids ( $M_{VS}$ ) was calculated to be  $1.28 \times 10^{-7}$  moles. The results of the experimental measured blanks are shown in Table 4. The mass of CO<sub>2</sub> recovered ranged from  $3.19 \times 10^{-5}$  to  $3.69 \times 10^{-5}$  moles. The mean experimental measured blank ( $MB_E$ ) was calculated to be  $3.44 \times 10^{-5}$  moles and was used in the calculations of the final calculated blank. The mass of the final calculated blank ( $CB_F$ ) was  $3.43 \times 10^{-5}$  moles CO<sub>2</sub>. This number was subtracted from the results of each sample to give the final mass of recoverable CO<sub>2</sub>.

### ADSORPTION

The adsorption experiments were performed five times with each sample. The results are shown in Tables 5 and 6. The CO<sub>2</sub> recovery was reproducible. Observed variation did not

correlate with variations in relative humidity. Table 5 displays the calculated mass of adsorbed CO<sub>2</sub> in mg per 20g (sample size) in each of the 5 runs of all samples. Mean recovered CO<sub>2</sub> ranged from 0.69 to 1.34 mg of CO<sub>2</sub>. Table 6 displays the calculated mass of adsorbed CO<sub>2</sub> in mg of CO<sub>2</sub> adsorbed per kg of ash. Recovered CO<sub>2</sub> ranged from 34.3 to 67.2 mg of CO<sub>2</sub> per kg of ash. The mean and standard deviation are shown in each table for all samples.

Figures 4-9 show a typical bake-off curve for each sample. The CO<sub>2</sub> concentrations are given in ppm which is equivalent to μmol of CO<sub>2</sub>/mol of nitrogen purge gas. The initial peak represents CO<sub>2</sub> in the headspace and larger pores of the ash. The concentration of the initial peak was significantly higher than atmospheric CO<sub>2</sub> concentration levels. This likely represents weakly adsorbed CO<sub>2</sub> and the head space and larger pores of the ash. The second peak is representative of strongly adsorbed CO<sub>2</sub> that was removed when heat is applied to the sample and gas within micro pores that may take longer to degas. The inset of each graph shows an enlargement of the second peak. The scales range from 10 to 210 ppm. The range is scale tends to make the graphs with lower ppm look like the sample has a longer baking off period. Each sample was baked off to a concentration lower than 1 ppm within 2 hours.

**Table 3. Specific surface area measurements**

<b>Volcano</b>	<b>Sample</b>	<b>Diameter (mm)</b>	<b>Specific Surface Area (m<sup>2</sup>/g)</b>
<b>Redoubt</b>	<b>RDASH1</b>	0.125	0.897
<b>Spurr</b>	<b>MSASH1</b>	0.5-0.7	1.174
<b>Okmok</b>	<b>OKASH1</b>	2-3	0.539
	<b>OKASH2</b>	2-3	0.548
<b>Tungurahua</b>	<b>TASH1</b>	0.375	0.127
	<b>TASH2</b>	0.375	0.148

**Table 4. Experimental measured blank results**

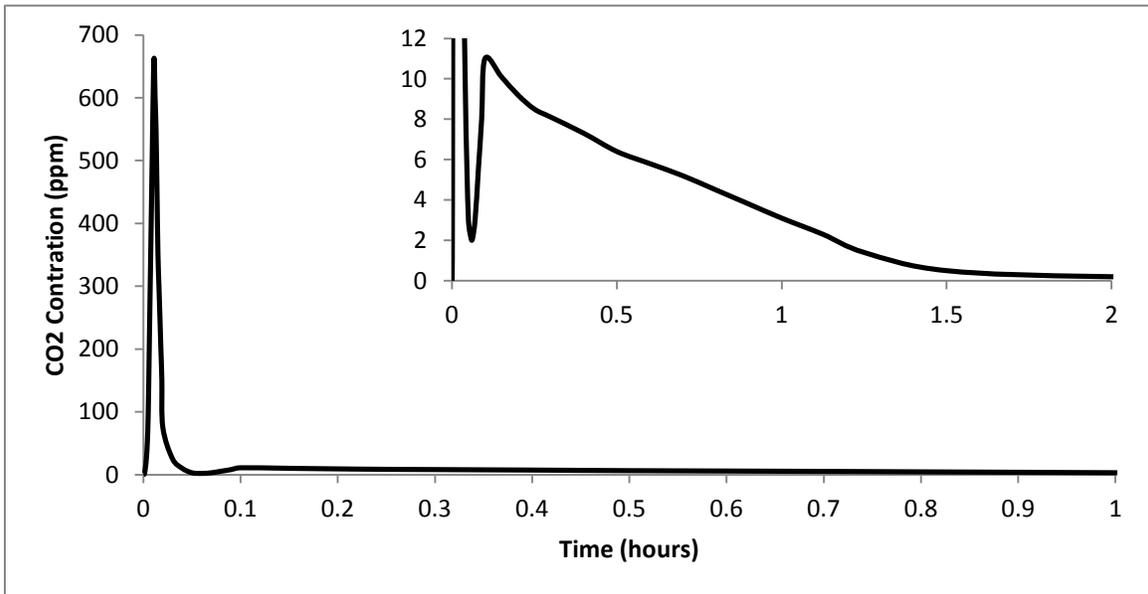
<b>Run #</b>	<b>CO<sub>2</sub> Adsorbed (moles)</b>
<b>1</b>	3.69 x 10 <sup>-5</sup>
<b>2</b>	3.57 x 10 <sup>-5</sup>
<b>3</b>	3.19 x 10 <sup>-5</sup>
<b>4</b>	3.32 x 10 <sup>-5</sup>
<b>Mean</b>	3.44 x 10 <sup>-5</sup>

**TABLE 5. Experimental results: mg of CO<sub>2</sub> per 20 g of sample**

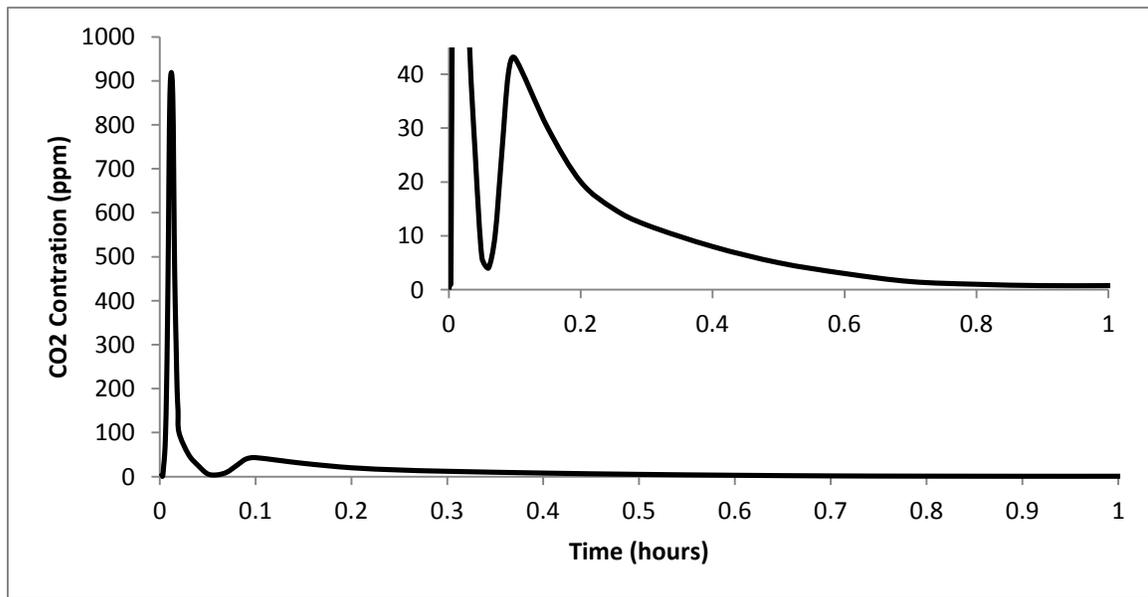
<b>Run #</b>	<b>RDASH1</b>	<b>MSASH1</b>	<b>OKASH1</b>	<b>OKASH2</b>	<b>TASH1</b>	<b>TASH2</b>
<b>1</b>	0.75	0.94	1.2	0.76	0.56	1.26
<b>2</b>	0.59	0.95	1.13	0.72	0.88	1.06
<b>3</b>	0.76	1.06	1.06	0.67	0.71	1.65
<b>4</b>	0.63	0.82	1.2	0.74	0.9	1.33
<b>5</b>	0.69	0.99	1.16	0.7	0.59	1.42
<b>Mean</b>	0.69	0.95	1.15	0.72	0.73	1.34
<b>Standard Deviation</b>	0.07	0.09	0.06	0.04	0.16	0.22

**TABLE 6. Experimental results: mg of CO<sub>2</sub> per kg of ash**

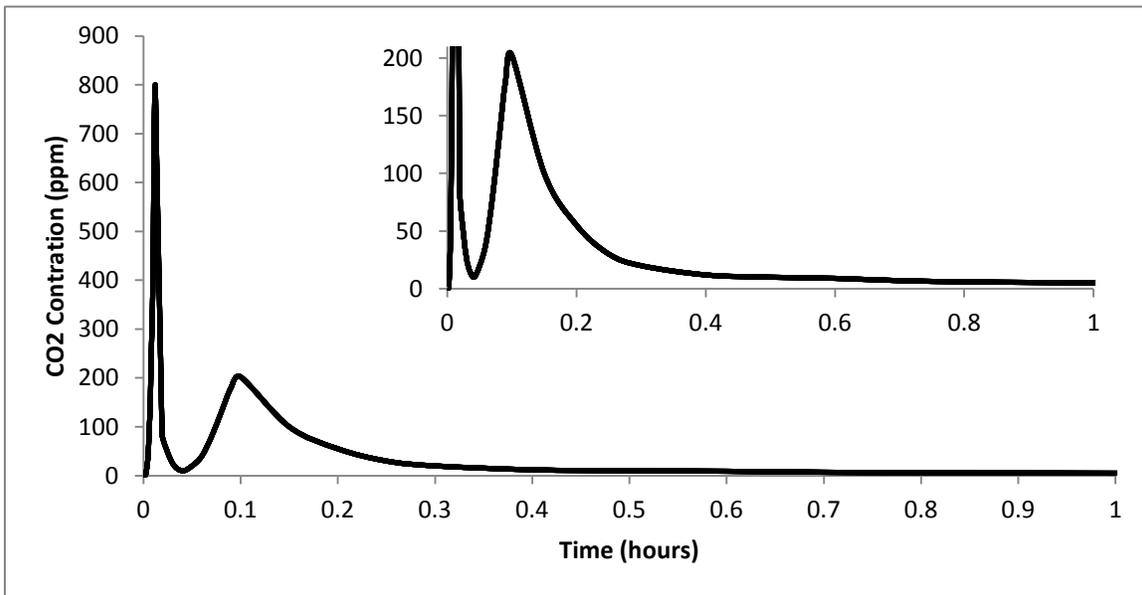
<b>Run #</b>	<b>RDASH1</b>	<b>MSASH1</b>	<b>OKASH1</b>	<b>OKASH2</b>	<b>TASH1</b>	<b>TASH2</b>
<b>1</b>	37.7	46.9	59.9	38.1	27.8	63
<b>2</b>	29.7	47.3	56.4	35.9	43.8	53
<b>3</b>	37.9	53	53.1	33.5	35.5	82.5
<b>4</b>	31.7	41.2	59.9	37	45.1	66.5
<b>5</b>	34.4	49.3	58.1	35	29.7	71
<b>Mean</b>	34.3	47.5	57.5	35.9	36.4	67.2
<b>Standard Deviation</b>	3.6	4.3	2.8	1.8	7.9	10.8



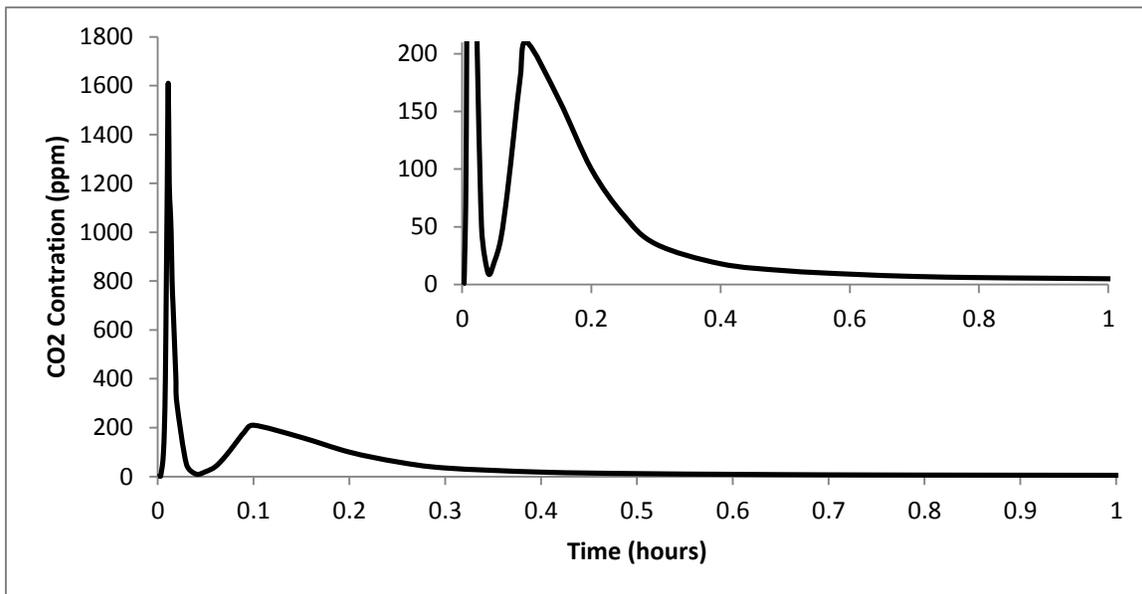
**FIGURE 4. Example bake off curve: RDASH1. Inset shows more detail on the CO<sub>2</sub> release while heated.**



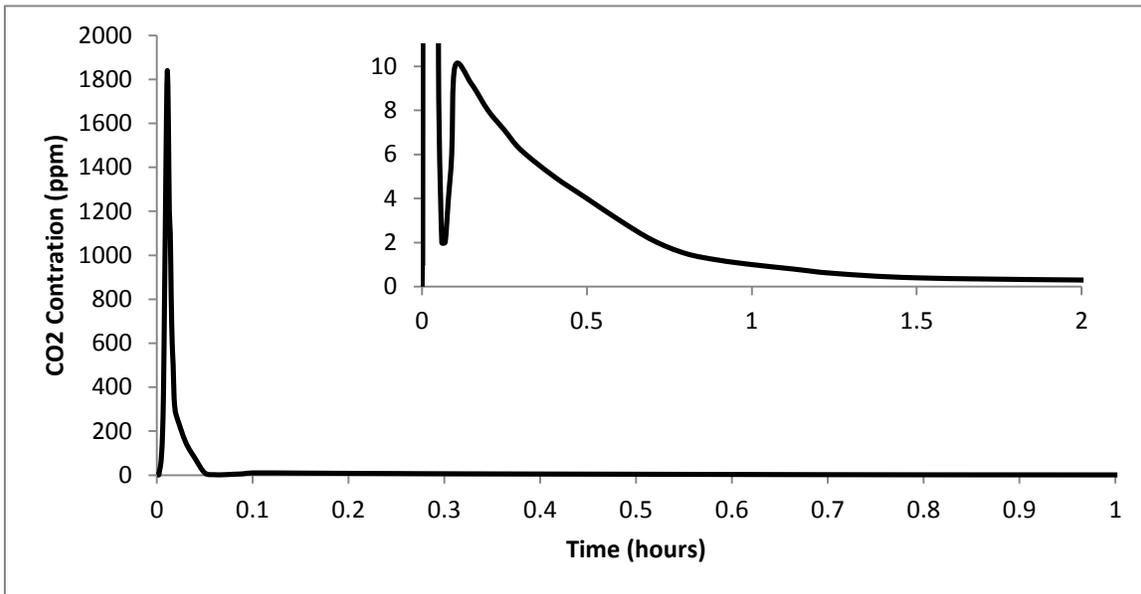
**FIGURE 5. Example bake off curve: MSASH1. Inset shows more detail on the CO<sub>2</sub> release while heated.**



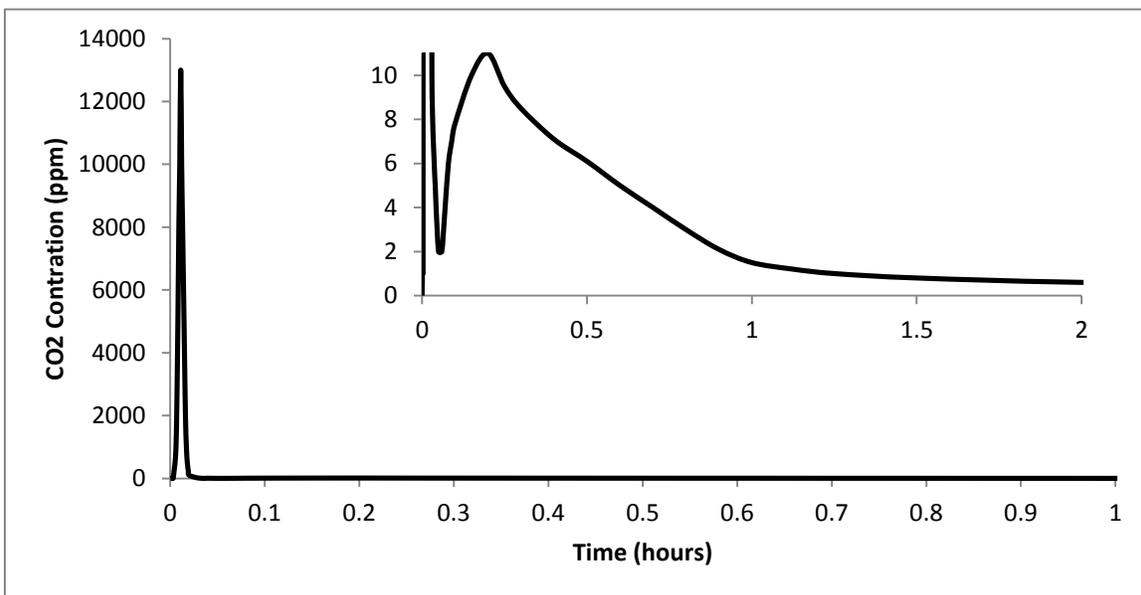
**FIGURE 6. Example bake off curve: OKASH1. Inset shows more detail on the CO<sub>2</sub> release while heated.**



**FIGURE 7. Example bake off curve: OKASH2. Inset shows more detail on the CO<sub>2</sub> release while heated.**



**FIGURE 8. Example bake off curve: TASH1. Inset shows more detail on the CO<sub>2</sub> release while heated.**



**FIGURE 9. Example bake off curve: TASH2. Inset shows more detail on the CO<sub>2</sub> release while heated.**

## ADSORPTION PER SPECIFIC SURFACE AREA

The mass of CO<sub>2</sub> adsorbed per m<sup>2</sup> of volcanic ash for each sample was calculated. The results are shown in Table 7. Adsorption ranged from 0.04 to 0.45 mg CO<sub>2</sub> per m<sup>2</sup> volcanic ash.

TASH2 had the highest adsorption of mg of CO<sub>2</sub> per kg of ash and the highest adsorption mg of CO<sub>2</sub> per m<sup>2</sup> of ash.

**Table 7. CO<sub>2</sub> adsorption: mg CO<sub>2</sub> per m<sup>2</sup> of ash**

<b>Volcano</b>	<b>Sample</b>	<b>CO<sub>2</sub> Adsorbed (mg CO<sub>2</sub>/m<sup>2</sup> ash)</b>
<b>Redoubt</b>	<b>RDASH1</b>	0.04
<b>Spurr</b>	<b>MSASH1</b>	0.04
<b>Okmok</b>	<b>OKASH1</b>	0.11
	<b>OKASH2</b>	0.07
<b>Tungurahua</b>	<b>TASH1</b>	0.29
	<b>TASH2</b>	0.45

## APPLICATION TO ERUPTIONS

Table 8 shows mass of ash and CO<sub>2</sub> released from eruptions, CO<sub>2</sub> adsorbed (mg of CO<sub>2</sub> adsorbed/kg of ash), total adsorption capacity, and estimated percent of released CO<sub>2</sub> adsorbed. The mean of the five runs was used to calculate mg of CO<sub>2</sub> per kg of ash. The eruption of volcano Redoubt released 0.75 x 10<sup>6</sup> metric tons CO<sub>2</sub> (Werner et al., 2013) and 55 x 10<sup>9</sup> kg of ash throughout the duration of the eruption (Wallace et al, 2013). RDASH1 adsorbed 34.3 mg of CO<sub>2</sub> per kg of ash, yielding a CO<sub>2</sub> adsorption capacity of 1,890 metric tons, equal to 0.25% of the CO<sub>2</sub> emitted from the volcano. The eruption of Mount Spurr released 85,000 tons of CO<sub>2</sub> (Doukas and Gerlach, 1995) and 36 x 10<sup>9</sup> kg of ash (McGimsey et al., 2001). MSASH1 adsorbed 47.5 mg of CO<sub>2</sub> per kg of ash, yielding a CO<sub>2</sub> adsorption capacity of 1,710 metric tons, equal to 2.01% of the CO<sub>2</sub> emitted from the volcano. For the samples lacking CO<sub>2</sub> measurements, the ratios of ash/CO<sub>2</sub> were calculated. This ratio used the mass of ash from the volcanos without CO<sub>2</sub> measurements to solve for CO<sub>2</sub> emissions matching the ratios from the volcanos with CO<sub>2</sub> data. The ratio is preferred to using estimates from similar eruptions. CO<sub>2</sub> emissions and mass of tephra released from the eruption of Mount Okmok were not measured. The calculations for CO<sub>2</sub> adsorption were performed using the mass of tephra from Volcano Kasatochi, 600 x 10<sup>9</sup> kg (Langmann et al., 2010). Volcano Kasatochi is located in the Aleutian Island chain and was erupting during the same time period. OKASH1 adsorbed 57.4 mg of CO<sub>2</sub> per kg of ash, yielding a CO<sub>2</sub> adsorption capacity of 34,400 tons. OKASH2 adsorbed 35.9 mg of CO<sub>2</sub> per kg of ash, yielding a CO<sub>2</sub> adsorption capacity of 21,500 tons. The calculations for adsorption percent were performed using the CO<sub>2</sub> emissions from Mount Spurr and Volcano Redoubt. Using the ash/CO<sub>2</sub> ratio from Mount Spurr, OKASH1 adsorbed 2.4% of emitted CO<sub>2</sub> and OKASH2 adsorbed 1.5% of emitted CO<sub>2</sub>. Using the ash/CO<sub>2</sub> ratio from Volcano Redoubt, OKASH1

adsorbed 0.42% of emitted CO<sub>2</sub> and OKASH2 adsorbed 0.26% of emitted CO<sub>2</sub>. Volcano Tungurahua erupted  $29 \times 10^9$  kg of ash (Bernard et al., 2016). TASH1 adsorbed 36.4 mg of CO<sub>2</sub> per kg of ash, yielding a CO<sub>2</sub> adsorption capacity of 1,060 tons. TASH2 adsorbed 67.2 mg of CO<sub>2</sub> per kg of ash, yielding a CO<sub>2</sub> capacity of 1,950 tons. CO<sub>2</sub> emissions from the eruption of Volcano Tungurahua were not measured. Using the ash/CO<sub>2</sub> ratio emissions from Mount Spurr, TASH1 adsorbed 1.5% of emitted CO<sub>2</sub> and TASH2 adsorbed 2.85% of emitted CO<sub>2</sub>. Using the ash/CO<sub>2</sub> ratio from Volcano Redoubt, TASH1 adsorbed 0.27% of emitted CO<sub>2</sub> and TASH2 adsorbed 0.49% of emitted CO<sub>2</sub>.

Several factors can reduce the impact of adsorption in the natural system. As deposited ash becomes wetted, a fraction of the adsorbed CO<sub>2</sub> is likely to desorb (Davidson et al., 2013), with the potential to minimize or negate the sink. Additional adsorption studies under wetted conditions will be required to quantify the impact. Volcanoes typically form at plate boundaries which are often adjacent to the ocean. Depending on the wind direction during the time of the eruption, the ejected pyroclastic material could settle in the ocean, resulting in a reduction of tephra available for adsorption of atmospheric CO<sub>2</sub>. The CO<sub>2</sub> emissions were measured used in calculations for the duration of the eruption. Volcanos do emit CO<sub>2</sub> before and after the eruption causing adsorption percent to decrease.

**TABLE 8. Volcanic eruption data and experimental results and calculations**

<b>Volcano</b>	<b>Sample</b>	<b>Ash Released (kg)</b>	<b>CO2 Released (t)</b>	<b>CO2 Adsorbed (mg/kg)</b>	<b>Adsorption Capacity (t)</b>	<b>Adsorption (%)</b>
<b>Redoubt</b>	<b>RDAS H1</b>	55 x 10 <sup>9</sup>	0.75 x 10 <sup>6</sup>	34.3	1890	0.25
<b>Spurr</b>	<b>MSAS H1</b>	36 x 10 <sup>9</sup>	85,000	47.5	1710	2.01
<b>Okmok</b>	<b>OKAS H1</b>	600 x 10 <sup>9*</sup>	8.2 x 10 <sup>6</sup> (Redoubt)	57.4	34400	0.42 (Redoubt)
	<b>OKAS H2</b>		1.42 x 10 <sup>6</sup> (Spurr)	35.9	21500	2.4 (Spurr)
<b>Tungurahua</b>	<b>TASH1</b>	29 x 10 <sup>9</sup>	0.36 x 10 <sup>6</sup> (Redoubt)	36.4	1060	0.27 (Redoubt)
	<b>TASH2</b>		0.0684 x 10 <sup>6</sup> (Spurr)	67.2	1950	1.5 (Spurr)
*= not measured						

## V. CONCLUSION

Adsorption of CO<sub>2</sub> was investigated on six volcanic ash samples of andesitic composition. Dry adsorption experiments were performed to collect and measure the amount of CO<sub>2</sub> adsorbed to the surface of the ash. Dry adsorption ranged from 34.3 to 67.2 mg of CO<sub>2</sub> per kg of ash. The volcanic ash from the eruptions investigated in this study were found to have an adsorption capacity that ranged from 1,060 to 34,400 tons of CO<sub>2</sub>, accounting for as little as 0.25 to as much as 2.85% of the CO<sub>2</sub> released by the eruption. Limitations in this research do exist. The lack of specific information on mass of CO<sub>2</sub> emissions and mass of ejected ash limited accuracy of calculations. Additionally, measurements on wetted ash should be conducted.

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