

**Indonesian volcanic  
emissions**

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# Atmospheric transport and deposition of Indonesian volcanic emissions

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## Abstract

A regional climate model study has been performed to investigate the transport and atmospheric loss rates of emissions from Indonesian volcanoes and the sensitivity of these emissions to meteorological conditions and the solubility of the released emissions. Two experiments were conducted: 1) volcanic sulfur released as primarily SO<sub>2</sub> and oxidation to SO<sub>4</sub><sup>2-</sup> determined by considering the major tropospheric chemical reactions; and 2) PbCl<sub>2</sub> released as an infinitely soluble passive tracer. The first experiment was used to calculate SO<sub>2</sub> loss rates from each active volcano resulting in an annual mean loss rate for all volcanoes of  $1.1 \times 10^{-5} \text{ s}^{-1}$ , or an e-folding rate of approximately 1 day. SO<sub>2</sub> loss rate was found to vary seasonally, be poorly correlated with wind speed, and uncorrelated with temperature or relative humidity. The variability of SO<sub>2</sub> loss rates is found to be correlated with the variability of wind speeds, suggesting that it is much more difficult to establish a “typical” SO<sub>2</sub> loss rate for volcanoes that are exposed to inconsistent winds. Within an average distance of 69 km away from the active Indonesian volcanoes, 53% of SO<sub>2</sub> is lost due to conversion to SO<sub>4</sub><sup>2-</sup>, 42% due to dry deposition, and 5% is lost due to lateral transport away from the dominant direction of plume travel. The solubility of volcanic emissions in water is shown to have a major influence on their atmospheric transport and deposition. High concentrations of PbCl<sub>2</sub> are predicted to be deposited near the volcanoes while volcanic S travels further away until removal from the atmosphere primarily via the wet deposition of H<sub>2</sub>SO<sub>4</sub>. The ratio of the concentration of PbCl<sub>2</sub> to SO<sub>2</sub> is found to exponentially decay at increasing distance from the volcanoes. The more rapid removal of highly soluble species should be considered when making observations of SO<sub>2</sub> in an aged plume and relating this concentration to other volcanic species. An assumption that the ratio between the concentrations of highly soluble volcanic compounds and S within an aged plume is equal to that observed in fumarolic gases will result in an overestimation of the atmospheric concentration of highly soluble species.

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## 1. Introduction

Volcanic emissions can have significant environmental effects on local, regional, and global scales dependent on how far the emissions are transported away from source prior to deposition. Characteristics of emissions, such as chemical and physical properties (including solubility and size), as well as environmental factors, i.e. the height at which emissions are released, wind speed, and precipitation, all influence transport.

Volcanic emissions can be released continuously by passive degassing or diffusive eruptions. Emissions can also be released sporadically by more violent, and short-lived, eruptions. [Andres and Kasgnoc \(1998\)](#) have calculated that 99% of volcanic SO<sub>2</sub> is released continuously, while only 1% is released during sporadic eruptions. The most violent of the sporadic eruptions can inject volcanic emissions into the stratosphere: there is generally at least one such stratosphere-reaching eruption every three years ([Simkin and Siebert, 1994](#)). Stratosphere-reaching eruption clouds generally cause global surface cooling for months up to a few years by sulfate aerosol (SO<sub>4</sub><sup>2-</sup>) backscattering of incoming shortwave solar radiation (e.g. [Textor et al., 2003](#)). Compared with stratosphere-reaching eruptive emissions, volcanic emissions released into the troposphere are rapidly deposited locally and regionally. Tropospheric volcanic emissions can have a significant atmospheric impact because such emissions are frequently released continuously for long periods of time, and because volcanoes are often at elevations above the planetary boundary layer, allowing those emissions to remain in the troposphere longer than, for example, most anthropogenic S emissions. As an example of the relative significance of non-eruptive volcanic degassing, such sources may be responsible for 24% of the total annual mean direct radiative top-of-atmosphere forcing ([Graf et al., 1997](#)).

Volcanic emissions are primarily H<sub>2</sub>O, followed by CO<sub>2</sub>, SO<sub>2</sub>, HCl, and other compounds (e.g. [Bardintzeff and McBirney, 2000](#)). Volcanic SO<sub>2</sub> has been the most monitored volcanic emission because the concentration of SO<sub>2</sub> within a volcanic plume is typically orders of magnitude greater in concentration than what is found in background

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ambient air. For the past few decades the majority of volcanic SO<sub>2</sub> observations have been performed with the Correlation Spectrometer (COSPEC), which measures the flux of emitted SO<sub>2</sub> (e.g. Stoiber et al., 1983). The (relatively) large number of published measurements of volcanic SO<sub>2</sub> fluxes is a useful tool for assessing the impact of volcanoes on the atmosphere because SO<sub>2</sub> is an environmentally important gas. SO<sub>2</sub> is readily converted, within days, to SO<sub>4</sub><sup>2-</sup> aerosol. SO<sub>4</sub><sup>2-</sup> is climatically significant (as described above when released explosively) and is a main component of acid rain.

Methods for observing tropospheric volcanic emissions include ground-based remote sensing, fumarolic gas sampling, and plume particle sampling. These techniques have contributed successfully to an improved understanding of the variations in time and between different volcanoes of emission compositions and strengths and, to a lesser extent, about processes occurring within volcanic plumes. There are, however, limitations to what can be accomplished in the field. For example, ground-based remote sensing measurements of volcanic SO<sub>2</sub> fluxes over time at one volcano can be used to observe changes in volcanic activity as an eruption prediction tool in conjunction with other volcano monitoring techniques (e.g. at Montserrat; Young et al., 2003). Remote sensing observations can detect that there is a change in the measured SO<sub>2</sub> flux, but cannot determine what observed variations are due to changes in the volcano itself and what are due to changing meteorological conditions. It can also be very difficult to determine what atmospheric processes are responsible for observed changes within a volcanic plume. One method of studying the loss of volcanic emissions from the atmosphere is to observe the variation of SO<sub>2</sub> concentration within a plume as the emissions move away from a volcano. A field-based method of characterizing this is to make measurements of atmospheric SO<sub>2</sub> at two distances away from a volcano, and to then relate these observations. SO<sub>2</sub> can be lost due to oxidation to SO<sub>4</sub><sup>2-</sup> or dry deposition, or it can appear to be lost due to lateral transport out of the observed plume. Ground-based remote sensing observations can measure the rate at which SO<sub>2</sub> is lost, but cannot measure to what extent each of the potential loss mechanisms have contributed to the SO<sub>2</sub> loss.

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Atmospheric chemistry modeling can be a useful tool to study processes occurring in the vicinity of active volcanoes that are difficult to measure directly. For example, modeled volcanic emission transport can be analyzed in light of variable meteorological conditions while the volcanic emissions are held constant. This removes the inherent natural variability of volcanic emission rates, so as to learn about what variations in atmospheric transport are due to changing atmospheric conditions rather than due to changes in the volcanic activity. Modeling can also be used to calculate what portion of  $\text{SO}_2$  lost from the atmosphere at increasing distances from active volcanoes is due to each of the mechanisms described above. Model experiments can further be used to study the transport and deposition patterns of volcanic emissions other than  $\text{SO}_2$ . Volcanic  $\text{SO}_2$  flux measurements using COSPEC are typically performed from distances of up to 30 km away from volcanic craters (for example at Mt. Etna; Weibring et al., 2002). Measurements performed at these distances are commonly used to estimate the flux rates of other volcanic compounds “X” by relating the observed concentration of  $\text{SO}_2$  in the plume to the ratio of “X” to total S found in fumarolic gases. This method assumes that the ratio of the concentrations of “X” to  $\text{SO}_2$  remains constant from the time the emissions are released until the plume is measured. This technique has been used, for example, to estimate the annual flux of metals from volcanoes (Hinkley et al., 1999) and to constrain the flux balances of elements at subduction zones (Hilton et al., 2002). The assumption of a steady ratio of  $[\text{X}]/[\text{SO}_2]$  remains a subject of uncertainty, however. Pyle and Mather (2003), for example, have shown that  $[\text{Hg}]/[\text{SO}_2]$  ratios can vary by an order of magnitude dependent on the type of volcanic activity (passively degassing vs. explosively erupting). The ratio of  $[\text{X}]/[\text{SO}_2]$  can vary not only dependent on the type of volcanic activity, but can also vary in time if the two species are removed at different rates from the plume. Atmospheric chemistry modeling can be a useful tool to study the transport and deposition of multiple chemical species and how they behave relative to  $\text{SO}_2$ . This approach can be used to gain insight onto how reasonable it is to relate observations of  $\text{SO}_2$  concentrations in an aged volcanic plume to other compounds.

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This paper describes a regional atmospheric chemistry modeling study that has been performed to address the two questions: 1) How do variable meteorological conditions influence volcanic SO<sub>2</sub> concentration in the atmosphere and SO<sub>2</sub> loss rates? and 2) How do the transport and deposition patterns of other volcanic compounds relate to SO<sub>2</sub>? Indonesia has been chosen as the region for study to address these two questions because it is the region of the world with the largest number of historically active volcanoes and the region has a relatively continuous emission history with 4/5 of the volcanoes with dated eruptions having erupted this past century (Simkin and Siebert, 1994).

## 2. Experimental setup

The regional atmospheric chemistry model REMOTE (Regional Model with Tracer Extension) (Langmann, 2000) has been used to simulate meteorological conditions for the year 1985, a climatologically “normal” year, i.e. neither “El Niño” nor “La Niña”. REMOTE combines the physics of the regional climate model REMO 5.0 with tropospheric chemical equations for 63 chemical species. The physical and dynamical equations in the model (Jacob, 2001) are based on the regional weather model EM/DM of the German Weather Service (Majewski, 1991) and include parameterizations from the global ECHAM 4 model (Roeckner, 1996). The chemical tracer transport mechanisms include horizontal and vertical advection (Smolarkiewitz, 1983), convective up- and down-draft (Tiedtke, 1989), and vertical diffusion (Mellor and Yamada, 1974). Trace species can undergo chemical decay in the atmosphere or can be removed from the atmosphere by wet and dry deposition or transport out of the model boundaries. Dry deposition is dependent on friction velocities and ground level atmospheric stability (Wesley, 1989). Wet deposition is dependent on precipitation rate and mean cloud water concentration (Walcek and Taylor, 1986). 158 gasphase reactions from the RADM II photochemical mechanism (Stockwell et al., 1990) are included. 43 longer-lived chemical species are treated as predicted species and 20 shorter-lived ones as diagnosed species.

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The model was applied with 20 vertical layers of increasing thickness between the Earth's surface and the 10 hPa pressure level (approximately 23 km). Analysis data of weather observations from the European Centre for Medium-Range Weather Forecasts (ECMWF) were used as boundary conditions every 6 h. The physical and chemical state of the atmosphere was calculated every 5 min. Background concentrations of 39 species (Chang et al., 1987), including SO<sub>2</sub>, SO<sub>4</sub><sup>2-</sup>, O<sub>3</sub>, and H<sub>2</sub>O<sub>2</sub>, were specified at the lateral model boundaries. The model domain covers Indonesia and Northern Australia (91° E–141° E; 19° S–8° N) with a horizontal resolution of 0.5° (approximately 53 km in longitude and 55 km in latitude) with 101 grid points in longitude and 55 grid points in latitude. Two experiments were performed: a) "S Experiment" – volcanic S was released as primarily SO<sub>2</sub> that underwent oxidation to SO<sub>4</sub><sup>2-</sup> following the major tropospheric chemical reactions and b) "PbCl<sub>2</sub> Experiment" – PbCl<sub>2</sub> released as an infinitely soluble passive tracer.

## 2.1. Emission inventory

An annual inventory was established to represent maximum potential volcanic emissions within the modeled region of Indonesia. Over the past century, from 1900 to 1993, 63 volcanoes in Indonesia are known to have erupted and 32 additional volcanoes have degassed passively, for a total sum of 95 active volcanoes (Simkin and Siebert, 1994). The inventory established for this work contains both continuous eruptive and passive degassing and sporadic eruptive volcanic emissions. Continuous emissions were taken from Nho et al. (1996) as this work provides the maximum published estimate of SO<sub>2</sub> emissions from the Indonesian volcanoes (Table 1: 1600 Gg SO<sub>2</sub>/yr released non-eruptively; 1900 Gg SO<sub>2</sub>/yr eruptively; for a sum of 3500 Gg SO<sub>2</sub>/yr continuous emissions (which is equivalent to 1750 Gg (S)/yr)). The continuous emissions were divided evenly amongst the 95 active volcanoes. This is the most reasonable assumption we could make, despite the fact that emission rates of volcanoes are highly variable in time and between different volcanoes, because only at a few of the active Indonesian volcanoes are routine SO<sub>2</sub> flux measurements performed. It would have been less

reasonable to have scaled the emission flux estimates for individual volcanoes based on the small number of available measurements for the active volcanoes. The result of dividing the continuous emissions evenly between all of the active volcanoes is a mean continuous SO<sub>2</sub> flux of 36.8 Gg SO<sub>2</sub>/yr (100 Mg SO<sub>2</sub>/day) for each volcano.

5 An estimate of the sporadic eruptive volcanic emissions for the region was established for this work using the [Simkin and Siebert \(1994\)](#) catalog of volcanic activity. [Simkin and Siebert \(1994\)](#) provide a compilation of the best known estimates of the date and strength for all of the known volcanic activity on Earth. Each volcanic eruption is assigned a volcanic explosivity index (VEI) strength which is an indicator of the  
10 explosiveness of a volcanic event ([Newhall and Self, 1982](#)). To assemble the sporadic emissions inventory, all of the eruptions recorded in the catalog during the last century (1900–1993) for each active Indonesian volcano were summed. An index estimating the amount of SO<sub>2</sub> released due to each VEI has been developed by [Schnetzler et al. \(1997\)](#), the volcanic sulfur index (VSI). We multiplied the total number of eruptions of  
15 each VEI by the maximum amount of SO<sub>2</sub> released by arc volcanoes suggested by the VSI. The SO<sub>2</sub> flux resulting from this multiplication was then divided by the 93 years of the record to generate an annual mean emission estimate. Averaging over 93 years removes some of the high natural short-term variability of volcanic activity. These calculations indicate 290 Gg SO<sub>2</sub>/yr released sporadically by the Indonesian volcanoes  
20 – a sum of sporadic and continuous volcanic emissions of 3800 Gg SO<sub>2</sub>/yr (which is equivalent to 1895 Gg(S)/yr) (Fig. 1). The estimated emission fluxes for the individual volcanoes correspond reasonably well with SO<sub>2</sub> flux measurements of Indonesian volcanoes (Table 2).

25 The emissions of each individual volcano were released into the model layer at the actual height of each volcano. The elevations of the volcanos range from 200 m (Riang Kotang) to 3805 m (Kerinci) corresponding to the first 12 model levels.

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## 2.2. Experiments

The “S Experiment” was conducted to observe the transport and deposition patterns of volcanic S. The volcanic emissions from the emission inventory were released into the model as 96% SO<sub>2</sub> and 4% SO<sub>4</sub><sup>2-</sup>. To describe the loss of the volcanic S from the atmosphere, SO<sub>2</sub> loss rate calculations have been performed on the results of the “S Experiment”. SO<sub>2</sub> loss rate is a function of the concentration of SO<sub>2</sub> at two locations within a volcanic plume, the distance between these two locations, and the time of travel from the first to the second location. The calculations have been performed in a manner intended to replicate the methodology of field measurements of tropospheric SO<sub>2</sub> loss rates at individual volcanoes (Oppenheimer et al., 1998).

SO<sub>2</sub> loss rate from the model results was calculated as follows: over a given time period (year or season), the mean wind direction of each gridbox containing a volcano “V” was used to define which of the 8 surrounding gridboxes the SO<sub>2</sub> was most likely to be transported to: “V+1”. This was repeated a second time to define the gridbox “V+2”, a distance of 55–200 km (average 121 km) away from the volcano. The mean column burden of SO<sub>2</sub> at “V” and “V+2” were then related following first order kinetics (Eq. 1).

$$\Phi_{t_1} = \Phi_{t_2} e^{k_1(t_2-t_1)} \quad (1)$$

where:

$\Phi$  = Column burden at given time [kg/m<sup>2</sup>]

$t_2-t_1$  = time to be transported from location 1 to 2 [s]

$k_1$  = SO<sub>2</sub> loss rate [s<sup>-1</sup>]

The mean wind speed and distance between the two gridboxes were used to calculate the amount of time for transport from “V” to “V+2”. The result of the calculation is the yearly or seasonal mean SO<sub>2</sub> loss rate “ $k_1$ ” for each volcano. Column burden of SO<sub>2</sub> was used in this calculation as opposed to single model level concentrations because column burden is a more accurate representation of the data obtained by

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ground-based COSPEC. Column burden is the total mass per area of the given species contained in the entire atmospheric vertical column (up to the top of the model, 10 hPa). For some volcanoes, the SO<sub>2</sub> loss rate calculation resulted in a negative or null value. A negative value indicates an increase in the concentration of SO<sub>2</sub> at “V+2” than at “V”. This can occur when the “V+2” grid box contains another volcano. A null value can occur when the wind direction is so variable that the emissions are predicted in the first step to be transported away from the grid box “V” and in the second step returned to it, for a net distance of 0. In both of these situations, the calculated SO<sub>2</sub> loss rates have been excluded from further consideration.

The “PbCl<sub>2</sub> Experiment” was conducted to observe the transport and deposition pattern of PbCl<sub>2</sub>, a highly soluble compound released by volcanoes in relatively large concentrations (e.g. Delmelle, 2003). PbCl<sub>2</sub> is not among the chemicals originally included in REMOTE, however, so we included PbCl<sub>2</sub> in the model as an infinitely soluble passive tracer. PbCl<sub>2</sub> (solubility = 0.99 g/100cc) (CRC Handbook, 1993) is very soluble, and not infinitely soluble, so the modeling assumption of infinite solubility will lead to an over-prediction of the solubility of PbCl<sub>2</sub>. The solubility is close enough, however, that we find it a reasonable proxy. The PbCl<sub>2</sub> is released as a passive tracer, and as such it is transported in the atmosphere and is removed from the atmosphere by wet and dry deposition processes, but it does not react to form other chemical species. The emission inventory was established for volcanic SO<sub>2</sub>, so to calculate a corresponding emission flux of PbCl<sub>2</sub> the emissions have been scaled to the ratio of Pb to S in Indonesian fumarolic gases (Table 3).

### 3. Results

The results of the “S Experiment” are presented first, followed by the SO<sub>2</sub> loss rates that have been calculated from these results. The results of the “PbCl<sub>2</sub> Experiment” are then presented.

### 3.1. "S Experiment" and calculated SO<sub>2</sub> loss rates

The modeled atmospheric distribution of volcanic S species is shown as annual mean column burden in Fig. 2 as a) SO<sub>2</sub>, b) SO<sub>4</sub><sup>2-</sup>, and c) total volcanic S (SO<sub>2</sub>+SO<sub>4</sub><sup>2-</sup>). The atmospheric concentration of SO<sub>2</sub> is much higher than that of SO<sub>4</sub><sup>2-</sup>, and dominates the sum of the two. The annual mean column burden of SO<sub>2</sub> ranges from 1.5–10 kg(S)/km<sup>2</sup> and SO<sub>4</sub><sup>2-</sup> from 0–1.5 kg(S)/km<sup>2</sup>. Qualitatively, both SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> show the highest concentrations near to the volcanoes, while away from the volcanoes the concentration decreases, with the dominant transport away from the volcanoes towards the east. Relatively high atmospheric concentrations of the S species is also seen at the northern boundaries of the figures. This is a result of the concentrations of SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> defined at the boundaries of the model domain and is not a result of the transport of volcanic S.

Volcanic S deposition is presented as a) the annual sum of the dry SO<sub>2</sub> deposition, b) dry + wet SO<sub>4</sub><sup>2-</sup> deposition, and c) the total volcanic S deposition as the sum of the two (Fig. 3). More than 99% of SO<sub>4</sub><sup>2-</sup> is deposited via wet deposition, so only the total SO<sub>4</sub><sup>2-</sup> deposition is shown. SO<sub>2</sub> is dry deposited in large concentrations close to the volcanoes, up to 3 Mg(S)/km<sup>2</sup>, but with almost no deposition away from the volcanoes. SO<sub>4</sub><sup>2-</sup>, in comparison, has a maximum annual deposition of only up to 1.25 Mg(S)/km<sup>2</sup>, with much more significant deposition away from the volcanoes. Most of the volcanic S is deposited as SO<sub>4</sub><sup>2-</sup> (83% of the total S deposition). There is an average annual sum of deposition over the entire modeled region of 45.6 kg(S)/km<sup>2</sup> SO<sub>2</sub> and 219.6 kg(S)/km<sup>2</sup> SO<sub>4</sub><sup>2-</sup>.

The SO<sub>2</sub> loss rates calculated from the model results ( $3.2 \times 10^{-7}$ – $4.1 \times 10^{-5}$  s<sup>-1</sup>) agree well in magnitude with SO<sub>2</sub> loss rates measured at individual volcanoes in other parts of the world ( $1.9 \times 10^{-7}$ – $5.4 \times 10^{-3}$  s<sup>-1</sup>) (Fig. 4) (Oppenheimer et al., 1998). There is a large variability in SO<sub>2</sub> loss rates measured at different volcanoes, and at Mt. Etna alone, SO<sub>2</sub> loss rates have been observed to vary over 3 orders of magnitude.

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Figure 5 shows a box plot of the bin wind speed over 1 m/s intervals plotted against SO<sub>2</sub> loss rates. The lower edge of the box represents the 25th percentile value and the upper edge the 75th. The height of each box shows the interquartile range for each season and is an indicator of the variability of the values. The line across the box indicates the median (50th percentile). Four outlayer values are shown as open circles and three extreme values as stars. The correlation between windspeed and SO<sub>2</sub> loss rate is weak but statistically significant ( $p < 0.01$ ;  $R^2 = 0.2$ ). There is a general trend of increasing wind speed associated with increased SO<sub>2</sub> loss rates as well as an increase in the variability of the SO<sub>2</sub> loss rates. Temperature and relative humidity, in contrast, demonstrate trivial and non-significant ( $R^2 < 0.02$ ) correlation with SO<sub>2</sub> loss rate.

SO<sub>2</sub> loss rates have been calculated for each season based on the monsoonal winds: winter monsoon (December–March); spring intermonsoon (April–May); summer monsoon (June–September); fall intermonsoon (October–November). SO<sub>2</sub> loss rates as a function of season are shown as a box plot in Fig. 6. Three outlayer values are shown as open circles and one extreme value as a star. Excluding the outliers and extremes, winter has the lowest variability and spring the highest. The mean seasonal SO<sub>2</sub> loss rates for all volcanoes vary between  $9.7 \times 10^{-6} \text{ s}^{-1}$  (spring) and  $1.3 \times 10^{-5} \text{ s}^{-1}$  (summer). A greater variability is demonstrated between individual volcanoes than by the seasonal means over all of the volcanoes.

Loss of volcanic SO<sub>2</sub> from the atmosphere can be accomplished via the dry deposition of SO<sub>2</sub> and by oxidation to SO<sub>4</sub><sup>2-</sup>. There can also be an apparent SO<sub>2</sub> loss due to lateral transport outside of the measured plume (in the field) or outside of the predicted transport route (in the calculations performed on the model results). The % of SO<sub>2</sub> lost due to dry deposition and oxidation was calculated by dividing the daily mean dry deposition of SO<sub>2</sub> and the annual mean column burden of SO<sub>4</sub><sup>2-</sup> in grid box “V” by the difference in column burden of SO<sub>2</sub> between locations “V” and “V+1”. The remaining lost SO<sub>2</sub> was attributed to lateral transport. The average for all volcanoes within an average of 69 km away from the volcanoes is 53% of SO<sub>2</sub> lost due to conversion to SO<sub>4</sub><sup>2-</sup>, 42% to dry deposition, and 5% lost due to lateral transport. This trend does not

continue at greater distances from the volcanoes. Between locations “V+1” and “V+2” (an average distance of 69–121 km from the volcanoes) the sum of the column burden of  $\text{SO}_4^{2-}$  and the daily dry deposition of  $\text{SO}_2$  is greater than the loss of  $\text{SO}_2$ .

### 3.2. “PbCl<sub>2</sub> Experiment”

5 The modeled atmospheric distribution of volcanic PbCl<sub>2</sub> is shown as annual mean column burden in Fig. 7. The annual mean column burden of PbCl<sub>2</sub> ranges from 0–3 g (Pb)/km<sup>2</sup>. Atmospheric PbCl<sub>2</sub> is found in greatest concentrations near to the volcanoes, with only slight easterly transport. The annual sum of the wet and dry PbCl<sub>2</sub> deposition is shown in Fig. 8. More than 99% of PbCl<sub>2</sub> is deposited via wet deposition,  
10 so only the sum of the two is presented. The PbCl<sub>2</sub> is deposited in concentrations of up to 2 kg (Pb)/km<sup>2</sup> with an average annual sum of 52 g (Pb)/km<sup>2</sup> of PbCl<sub>2</sub> deposited in the modeled region.

## 4. Model result verification

We will assess the quality of the modeling results by comparing the modeled S deposition with the concentration of S measured in peat core samples collected in the modeled region. Peat can serve as a historical record of atmospheric deposition for  
15 time periods of up to thousands of years. The peat areas of Indonesia may be particularly useful recorders of the deposition of volcanic emissions because of the large number of historical and modern active volcanoes in the vicinity of peat areas (Langmann and Graf, 2003). It has been suggested in several studies that anomalous, high concentrations of S and other chemicals including Pb in peat core samples (collected outside of Indonesia) may be due to volcanic deposition (e.g. Weiss et al., 1997; Roos-Barracough et al., 2002; Kylander et al., 2005). Within Indonesia, there are two main types of peat: ombrogenous and topogenous (Page et al., 1999). Ombrogenous peat  
20 receives nutrients only from atmospheric deposition while topogenous peat also re-

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ceives nutrients from groundwater. Ombrogenous peat is therefore more useful for interpreting the historical deposition of atmospheric compounds. In this work we have taken measurements of S in four ombrogenous peat areas in Indonesia for comparison with the modeled S deposition (Fig. 9; Table 4). In making this comparison, it is important to keep in mind that there are other natural sources of S additional to volcanoes, such as vegetation and sea spray, so this comparison can be only qualitative.

The average S of each sampled peat core was calculated by multiplying the average % S in each of the four peat sampling locations with the average peat dry bulk density ( $0.18 \text{ g/cm}^3$ ) given by Shimada et al. (2001). This value was multiplied by the minimum ( $1.7 \text{ mm/yr}$ ) and maximum ( $4.3 \text{ mm/yr}$ ) peat accumulation rates provided by Supardi et al. (1993), resulting in the presented range of values for the S deposition of each peat core. The average % S was calculated from 3–16 samples within each peat core. Each peat core had measurements of both total S as well as  $C_{14}$  ages, or were very near to another peat core where  $C_{14}$  age measurements were performed. S values from portions of the peat cores that were dated to be less than 150 years old were not included in the average as these S values may have been influenced by human activity. The modeled S deposition and the rate of S deposition measured in the peat core samples are of the same order of magnitude. The potential volcanic S contribution to the peat areas ranges from 6–72% of the S measured in the peat samples (Table 4). There is a relatively uniform concentration of volcanic S predicted to be deposited on all four peat areas ( $215\text{--}285 \text{ kg/km}^2 \text{ yr}$ ). This is because of the distance between the peat areas and the nearest volcanoes (minimum 153 km). It would be helpful to be able to compare the model results with a peat sample collected nearer to the volcanoes, but we have not been able to find such a sample. We find the agreement in scale to be a strong indication that the modeled deposition of the volcanic S is reasonable and feel confident in interpreting the model results for the transport and deposition of volcanic emissions.

## 5. Discussion

We will interpret the modeling results and discuss how these results can be used to address the two questions described above: 1) How do variable meteorological conditions influence volcanic SO<sub>2</sub> concentration in the atmosphere and SO<sub>2</sub> loss rates? and 2) How do the transport and deposition patterns of other volcanic compounds relate to SO<sub>2</sub>?

### 5.1. SO<sub>2</sub> loss rate

Temperature, relative humidity, and wind speed have been plotted against the relative % of SO<sub>2</sub> lost due to the dry deposition of SO<sub>2</sub>, oxidation to SO<sub>4</sub><sup>2-</sup>, and lateral transport outside of the predicted plume pathway to see if there is any correlation between variations in the meteorological conditions and the manner in which SO<sub>2</sub> is lost. No such correlation was found. There is an observable seasonal cycle of SO<sub>2</sub> loss rates with the lowest loss rates in spring and the highest in summer. The only seasons with outlayers and extreme values are summer and winter – the monsoon seasons – which are distinguished by strong winds. The model results suggest, albeit weakly, that there may be a relationship between stronger winds and greater SO<sub>2</sub> loss rates. There is a stronger relationship revealed between stronger winds and greater variability of SO<sub>2</sub> loss rates.

The large variabilities of SO<sub>2</sub> loss rates measured at individual volcanoes have been attributed to variable atmospheric and plume conditions (Oppenheimer et al., 1998). The results of this study suggest a refinement of this assessment, in that the meteorological condition most significantly influencing the variability of SO<sub>2</sub> loss rates is wind speed. It may be more difficult to obtain a representative SO<sub>2</sub> loss rate for a given volcano that is susceptible to highly variable wind conditions, as opposed to a volcano that is exposed to more consistent winds. Further fieldwork-based research that considers variations in wind speed and apparent SO<sub>2</sub> loss rates may be able to form a more conclusive statement about the possible correlation between wind speed and

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SO<sub>2</sub> loss rates. If there is indeed such a relationship, it may be important to consider wind speed variations when making interpretations about changes in volcanic activity based on remote SO<sub>2</sub> measurements. Some variations in SO<sub>2</sub> flux observed over time at one volcano may be due to differences in the winds, as opposed to variations in the volcanic emissions.

## 5.2. Differences in transport and deposition patterns due to solubility

Both the atmospheric burden and deposition of Pb are three orders of magnitude less than that of S. In both experiments, deposition is relatively uniform with relation to distance from any given volcano and not very distinctive for individual volcanoes. This uniformity is a result of the assumption of an even distribution of the continuous volcanic emissions between the active volcanoes. PbCl<sub>2</sub> is rapidly deposited very close to the volcanoes, resulting in high local concentrations and a sharp decline in deposition at greater distances from the volcanoes. SO<sub>2</sub>, on the other hand, is much less soluble in rain than PbCl<sub>2</sub>. The less soluble SO<sub>2</sub> has some dry deposition, but is mostly transported away from the volcanoes prior to conversion to water-soluble SO<sub>4</sub><sup>2-</sup>. The SO<sub>2</sub> that is deposited, however, is deposited at heavier concentrations near to the volcanoes than the PbCl<sub>2</sub>. Because most of the SO<sub>2</sub> is converted to SO<sub>4</sub><sup>2-</sup> rather than deposited directly as SO<sub>2</sub>, there is a much less steep gradient of S deposition at increasing distance from the volcanoes compared with PbCl<sub>2</sub>, as well as a higher concentration of S deposition at greater distances from the volcanoes.

The influence of solubility on deposition patterns is illuminated by comparing the results of the two performed experiments (Fig. 10). The strong dependency of deposition rate on solubility has implications for the accurate extrapolation of measurements of SO<sub>2</sub> flux in aged volcanic plumes to other compounds. The further away from a volcano such measurements are made, the less accurate it is to assume that the concentration of volcanic SO<sub>2</sub> measured there has the same ratio to more soluble species as the ratio measured in fumarolic gases.

The ratio of [Pb]/[S] in the air decreases with increasing distance from the volcanoes

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as the  $\text{PbCl}_2$  is deposited (Fig. 11). Figure 11 is a box plot with the same specifics as for Figs. 5 and 6. Four outlayer values are shown as open circles at location “V”. The interquartile range increases at greater distance from the volcanoes indicating that the variability of the  $[\text{PbCl}_2]/[\text{SO}_2]$  ratio is growing at greater distances from the volcanoes.

- 5 The median  $[\text{PbCl}_2]/[\text{SO}_2]$  ratio decreases exponentially at greater distances from the volcanoes with the mean exponential rate of decay of the  $[\text{PbCl}_2]/[\text{SO}_2]$  ratio based on these three distances being  $y=106.5e^{-0.002x}$

where:

$$y = [\text{PbCl}_2]/[\text{SO}_2] \text{ (}\mu\text{g/g)}$$

- 10  $x = \text{distance from volcanoes (km)}$ .

The mean  $[\text{PbCl}_2]/[\text{SO}_2]$  ratio at the three distances are: “V” = 107.7; “V+1” = 89.3; and “V+2” = 83.2  $\mu\text{g/g}$ .

- 15 Based on this mean rate of decay, we estimate that calculations (e.g. based on COSPEC measurements) which assume a constant  $[X]/[S]$  ratio as found in fumarolic gases will result in a 6% overestimation of the atmospheric concentration of highly soluble species at 30 km distance away from the volcano. The overestimation grows at further distances from the volcano.

## 6. Conclusions

- 20 This study demonstrates that realistic modeling of volcanic emissions can lead to an improved understanding of the atmospheric processes occurring in the vicinity of active volcanoes. The results of the study demonstrate that  $\text{SO}_2$  loss rates are weakly correlated with wind speed and uncorrelated with relative humidity or temperature and that there is no correlation between these three meteorological phenomena and the relative amount of  $\text{SO}_2$  lost due to the dry deposition of  $\text{SO}_2$ , conversion to  $\text{SO}_4^{2-}$ , or lateral transport. A relationship is shown between increased wind speed and increased variability of  $\text{SO}_2$  loss rates. We recommend that further fieldwork-based research be
- 25 conducted to explore the possible relationship between wind speed and apparent  $\text{SO}_2$

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loss rates as variations in wind speed might lead to changes in SO<sub>2</sub> loss rates independent of a change in the state of volcanic activity.

The solubility of volcanic emissions is shown to control if they are deposited near to the volcanoes or transported prior to deposition. Highly soluble species such as PbCl<sub>2</sub> have high deposition rates near to the volcanoes while the relatively insoluble SO<sub>2</sub> is transported away from the volcanoes until it is oxidized to SO<sub>4</sub><sup>2-</sup> and then rapidly deposited. The ratio of [X]/[SO<sub>2</sub>], with “X” being a soluble species, decreases exponentially at greater distances from the volcanoes. We therefore recommend that the influence of different solubilities of volcanic species on atmospheric loss should be considered when relating measurements of atmospheric SO<sub>2</sub> to other volcanic emissions.

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**Table 1.** Estimates of Indonesian volcanic emissions.

Emission style	SO <sub>2</sub> flux (Gg/yr)	Reference
continuous (non-eruptive)	120	<a href="#">Hilton et al. (2002)</a>
continuous (non-eruptive)	210	<a href="#">Spiro et al. (1992)</a>
continuous (non-eruptive)	1600	<a href="#">Nho et al. (1996)</a>
continuous (eruptive)	1900	<a href="#">Nho et al. (1996)</a>
continuous (eruptive + non-eruptive)	3500	sum from <a href="#">Nho et al. (1996)</a> used in this study
sporadic (eruptive)	290	calculated for this study
continuous + sporadic (eruptive + non-eruptive)	2100–2600	<a href="#">Halmer et al. (2002)</a>
continuous + sporadic (eruptive + non-eruptive)	3790	this study

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**Table 2.** SO<sub>2</sub> emissions from individual Indonesian volcanoes.

Volcano	SO <sub>2</sub> flux from emission inventory (Gg/yr)	SO <sub>2</sub> flux from measurements (Gg/yr)	Reference
Bromo (Tengger Caldera)	47.4	5.1	<a href="#">Andres and Kasgnoc (1998)</a>
Galunggung	47.7	140.5	<a href="#">Bluth et al. (1994)</a>
Merapi	55.6	240.9	<a href="#">Andres and Kasgnoc (1998)</a>
		36.5	<a href="#">Dir. of Volcan. and Geol. Haz. Mit. of Indonesia (2005)</a>
		51.1	<a href="#">Andres and Kasgnoc (1998)</a>
Slamet	45.4	73.0	<a href="#">LeGuern (1982)</a>
		21.2	<a href="#">Nho et al. (1996)</a>
Tangkubanparahu	37.5	27.4	<a href="#">Andres and Kasgnoc (1998)</a>

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**Table 3.** Pb/S ratios in Indonesian volcanic gases.

Volcano	Pb/S ( $\mu\text{g/g}$ )	Reference
Merapi	420	<a href="#">Nho et al. (1996)</a>
Merapi	35	<a href="#">Symonds et al. (1987)</a>
Papandayan	280	<a href="#">Nho et al. (1996)</a>
Mean	245	the average of the above measurements was applied in this study
Global mean	190	<a href="#">Hinkley et al. (1999)</a>

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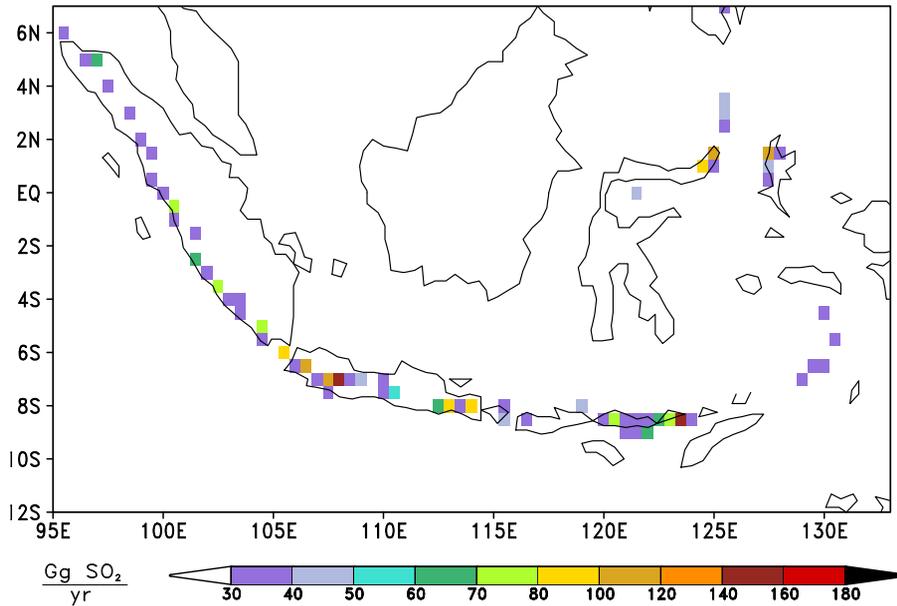
**Table 4.**

Comparison of modeled S deposition and peat core samples.

Sampling location	Distance to nearest volcano (km)	Measured S accumulation (kg/km <sup>2</sup> – yr)	Modeled S deposition (kg/km <sup>2</sup> – yr)	Volcanic S (%)	Reference (% S measured in peat core)
Riau	153	398–1006	285	28–72	<a href="#">Supardi et al. (1993)</a>
Batanghari River	160	1744–4412	264	6–15	<a href="#">Esterle and Ferm (1994)</a>
Tasek Bera	258	796–2012	215	11–27	<a href="#">Wüst and Bustin (2001)</a>
Sungai Seban-gau	396	428–1084	253	23–59	<a href="#">Weiss et al. (2002)</a>

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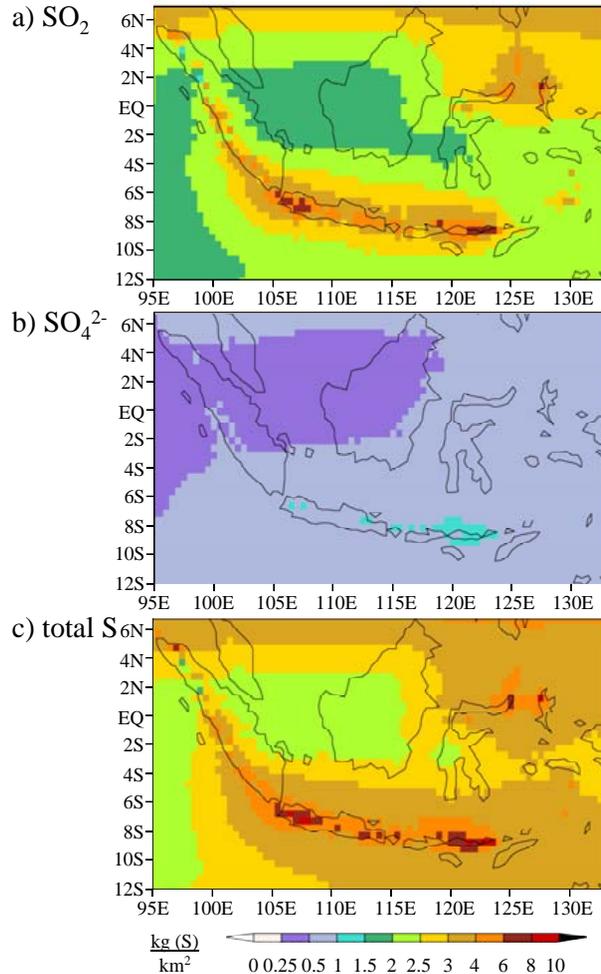
**Fig. 1.** Emission inventory of the annual sum of continuous (eruptive + passive) and sporadic (eruptive) volcanic SO<sub>2</sub> emissions.

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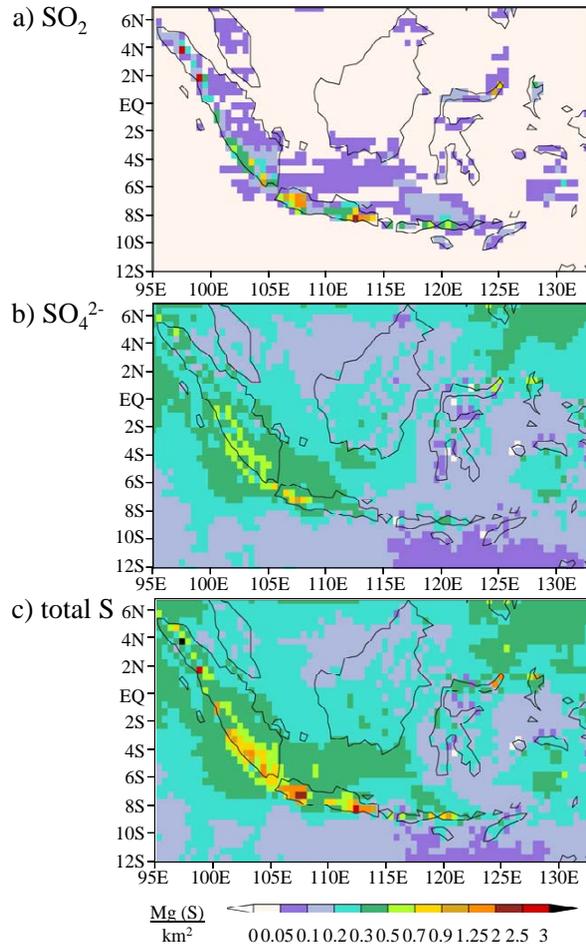


**Fig. 2.** Annual mean vertical column burden of (a) SO<sub>2</sub>, (b) SO<sub>4</sub><sup>2-</sup>, and (c) total S: SO<sub>2</sub>+SO<sub>4</sub><sup>2-</sup> for the “S Experiment”.

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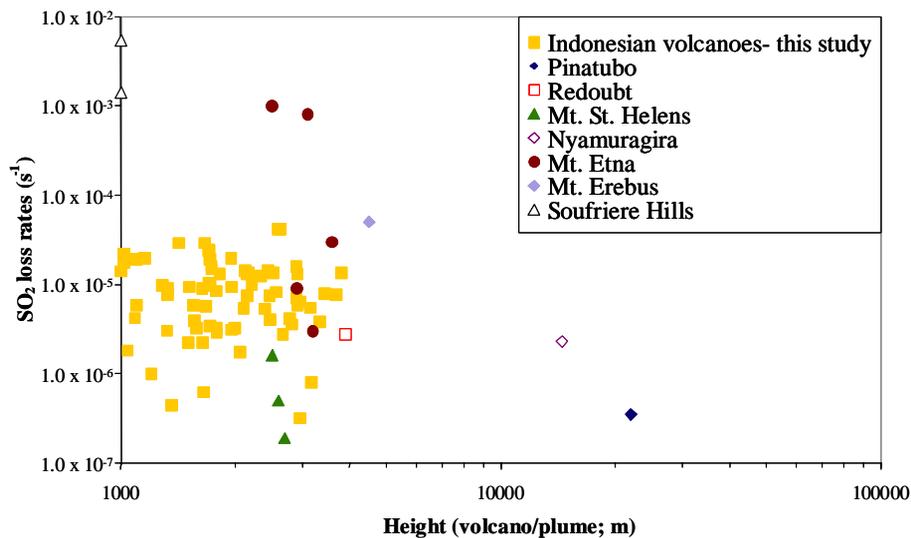


**Fig. 3.** Annual sum of the **(a)** dry SO<sub>2</sub> deposition, **(b)** dry + wet SO<sub>4</sub><sup>2-</sup> deposition, and **(c)** total S: dry SO<sub>2</sub> + dry + wet SO<sub>4</sub><sup>2-</sup> deposition for the “S Experiment”.

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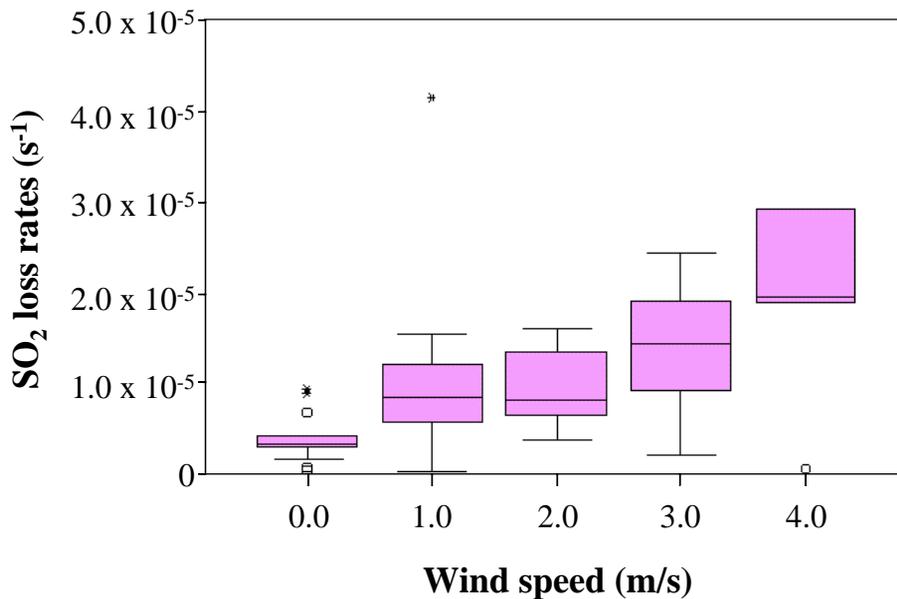
**Fig. 4.** Modeled SO<sub>2</sub> loss rates (yellow squares) are plotted against the actual height of each volcano and measured SO<sub>2</sub> loss rates from Oppenheimer et al. (1998) are plotted against the observed plume height.

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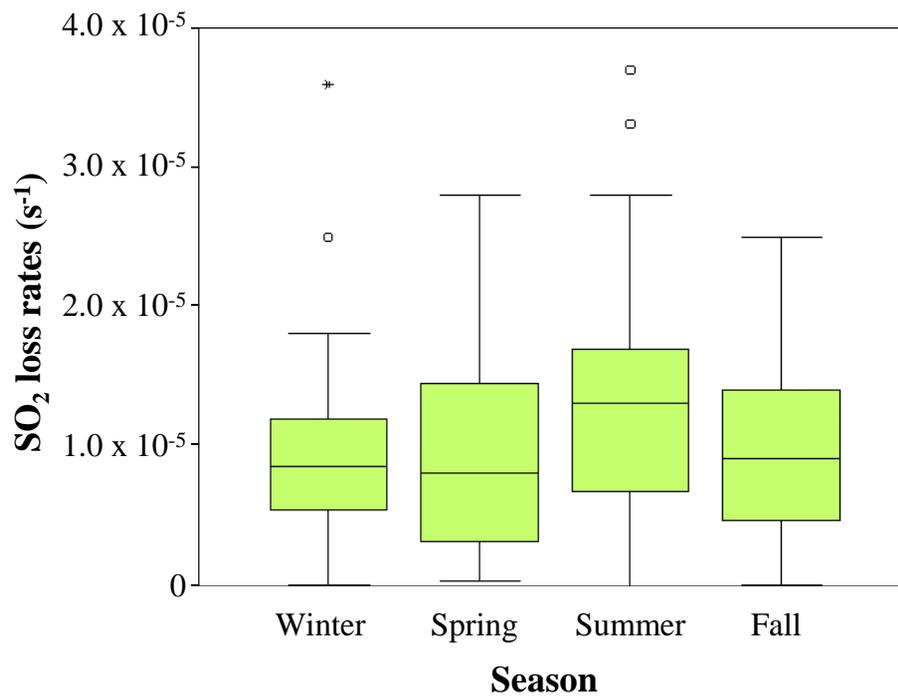
**Fig. 5.** Annual mean SO<sub>2</sub> loss rates for each modeled volcano plotted against the annual mean wind speed at the height of each volcano.

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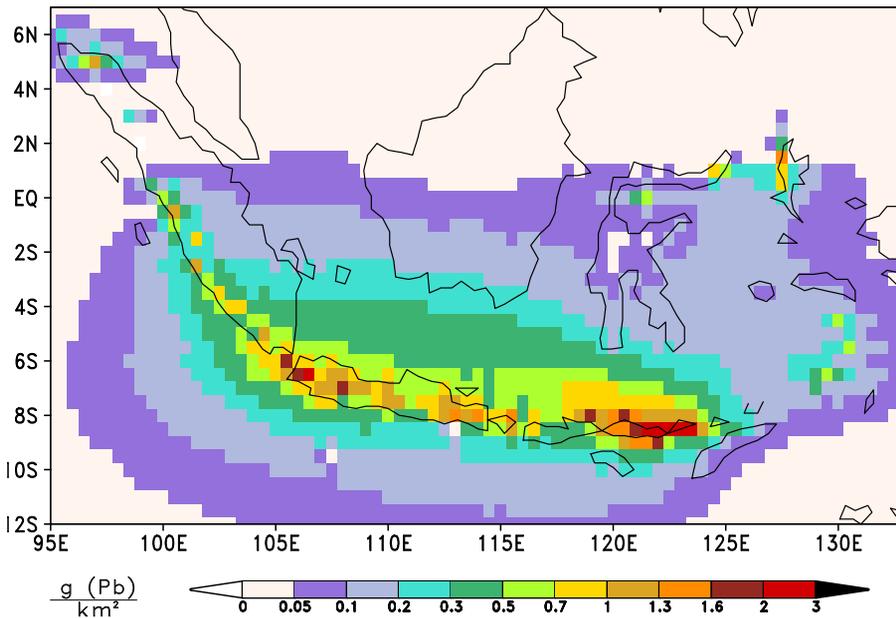
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**Fig. 6.** Seasonal SO<sub>2</sub> loss rates.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

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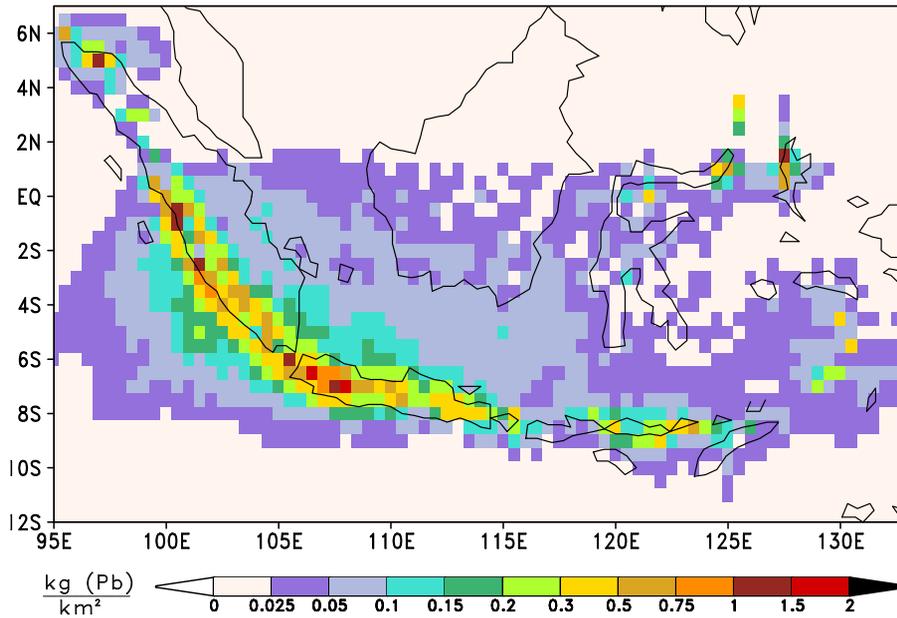
**Fig. 7.** Annual mean column burden of  $\text{PbCl}_2$  for the “ $\text{PbCl}_2$  Experiment”.

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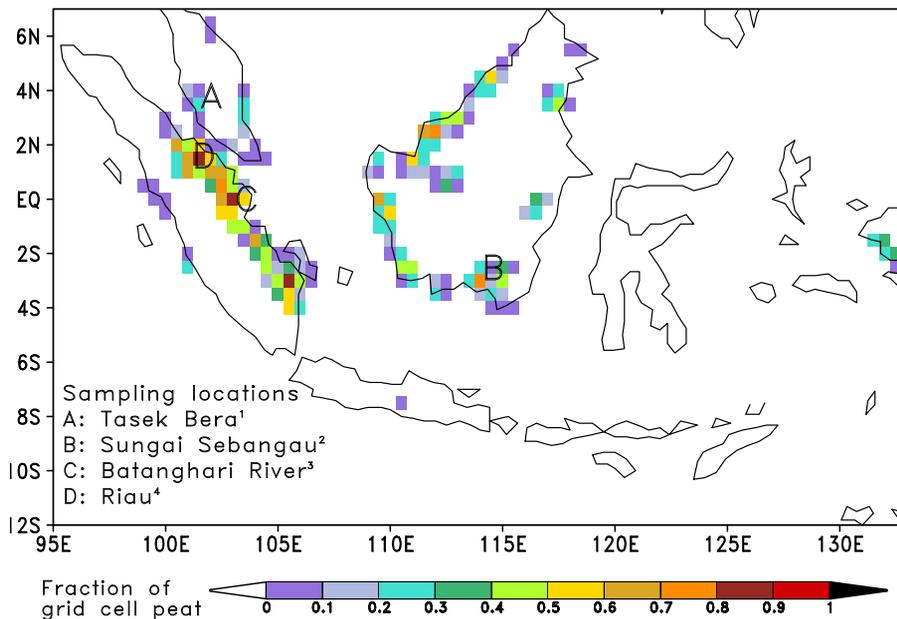
**Fig. 8.** Annual sum of dry + wet  $\text{PbCl}_2$  deposition for the "PbCl<sub>2</sub> Experiment".

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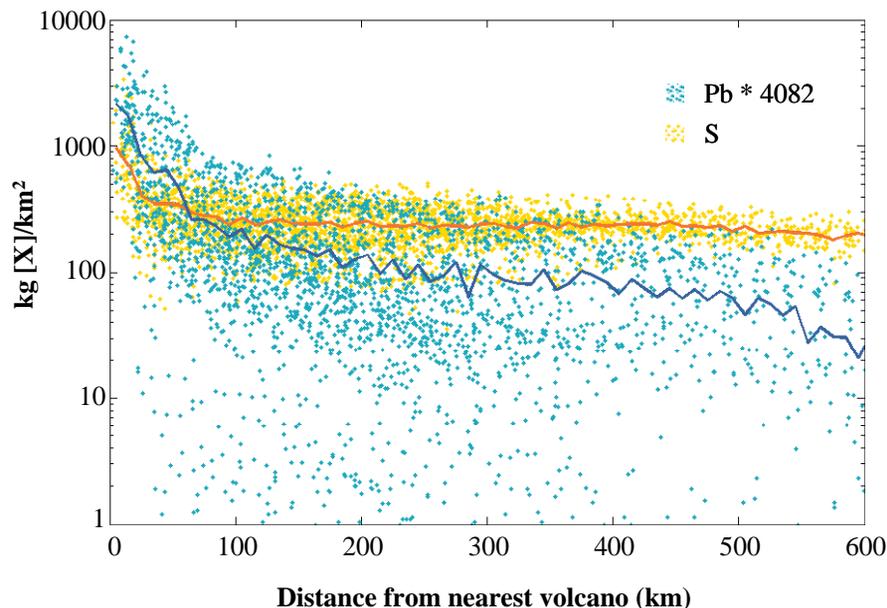
**Fig. 9.** Peat sampling locations on a map of the fractional peat coverage of the modeled region (after Heil et al., 2005). <sup>1</sup> Wüst and Bustin (2001); <sup>2</sup> Weiss et al. (2002); <sup>3</sup> Esterle and Fern (1994); <sup>4</sup> Supardi et al. (1993).

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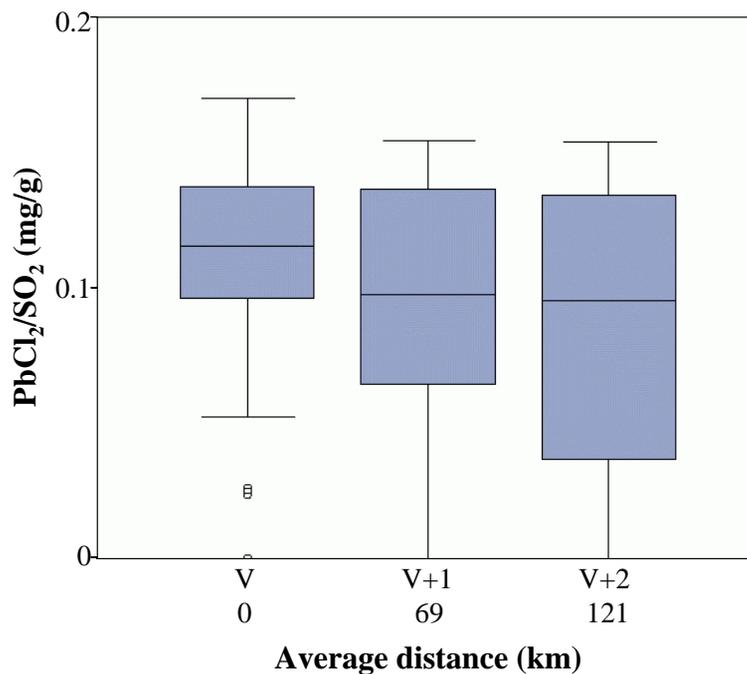
**Fig. 10.** Annual sum of total deposition for each modeled grid box as a function of the distance to the nearest volcano (“Passive experiment”: blue; “S experiment”: yellow). The solid lines (corresponding colors) are the bin mean over 10 km intervals.

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**Fig. 11.** Annual mean column burden of  $[\text{PbCl}_2]/[\text{SO}_2]$  for all volcanoes plotted against the mean distance from each volcano (km) for locations “V”, “V+1”, and “V+2”.

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