"Acid neutralization and sulphur retention in s-impacted andosols"

Delfosse, Thomas

Abstract
While Andosols have a proven capacity to buffer acid inputs, their long-term chemical response to elevated acid deposition remains poorly known. In this respect, the high anion retention capacity of Andosols constitutes a key parameter. Yet, the mechanisms involved in anion retention, especially sulphate, are still a matter of scientific debate. In this study, we report on the impacts of volcanogenic S and acid depositions on (i) the sulphate distribution and (ii) the processes involved in the neutralisation of the acid inputs, in two distinct soil series located downwind from Masaya volcano (Nicaragua), one of the world's largest natural source of SO2. The first series corresponds to weathered Eutric Andosols rich in allophanic constituents and the second series to weakly developed Vitric Andosols rich in volcanic glass. Long-term acid gas emission by Masaya volcano has led to important changes in the chemistry of the Andosols downwind. Sustained acid inputs have decreased the pH an...

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Acid Neutralization and Sulphur retention in S-impacted Andosols

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Part III

Environmental and Agronomical Issues
Chapter 9

Environmental and Agronomical Impacts of Volcanic Emissions

9.1 Improving critical load models for acid deposition

In order to investigate the potential impact of acidifying deposition generated by anthropogenic activities, the critical load approach has been developed in Europe during the 1980’s and 1990’s (Nilsson & Grennfelt, 1988). While volcanic emissions into the low atmosphere may result in strong acid inputs to local terrestrial ecosystems (Chapter 2), the critical load approach has never been applied in the affected areas. It is therefore relevant to query the extent to which the critical load approach may be applied to volcanic areas that are often located in tropical and developing countries.

Critical load is used to estimate ecosystem sensitivity quantitatively. This concept allows the mapping of areas sensitive to acid deposition, which in turn highlights the areas where acid deposition exceeds what soil, vegetation or surface waters can tolerate. Therefore, critical load links emissions and ecosystem effects. The critical load is defined as: ‘a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge’ (Nilsson & Grennfelt, 1988). This quantitative estimate may be compared with calculated, measured or projected sulphur and nitrogen acidifying deposition rates, and it is assumed that excess deposition above critical loads is likely to cause detrimental impacts to elements of the ecosystem.
Developing countries have different ecosystems and climate, and in these regions, relevant data are often incomplete or, simply lacking. Given the difficulties in application of the critical load concept to areas outside Europe and the global scale of the investigation, a simple methodology has been developed by Kuylenstierna (2001). Rather than calculating critical loads directly in the way it is carried out in Europe, the model consists in mapping relative ecosystem sensitivities for which preliminary values of critical loads are assigned. These are then compared to net acidic deposition values. Kuylenstierna (2001) used a proxy variable (base saturation or ‘BS’), which reflects the soil weathering rate. BS corresponds to the ratio of exchangeable base content to total cation exchange capacity. BS will tend to be high in soils with a high weathering rate, and low in soils with low weathering rate; other things being equal. It has the advantage of being a frequently measured variable, and it is possible to relate it to mapped soil types.

More information on weathering rates is therefore required to improve critical load estimates, especially in tropical ecosystems (Fumoto & Sverdrup, 2000). To estimate Al concentrations critical load models are generally based on the weathering rate of gibbsite. However, the main Al source of volcanic ash soils is not gibbsite, but volcanic glass, and the rate of weathering of this material is significantly higher than that of gibbsite (Shoji et al., 1993).

The weathering rate of volcanic soils containing volcanic glass could be estimated using the surface area of the sand fraction as an estimate of surface area of weatherable minerals (Fumoto et al., 2001). Ideally, the method should also account for the chemical composition and morphology of the minerals. This is best relevant to better assess and mitigate the impacts of acid deposition on poorly weathered volcanic soils such as the Vitric Andosols of the Masaya area that neutralise acid inputs mainly through mineral weathering (Chapter 3).

The solubility of Al is also one of the most influential variables for calculating critical loads. If recent Al solubility models incorporate the possible complexation of Al by organic matter, few models include the potential complexation of Al by F anions (Fumoto et al., 2001). Yet, in volcanic areas where soils may be exposed to intense F loading, formation of Al-F\textsubscript{X} complexes strongly dictates the mobility and solubility of Al (Chapter 5).

Many soils in the tropics are highly weathered. These acidic soils not only have low rates of weathering, but also may exhibit a high sulphate adsorption capacity. Sulphate adsorption mainly releases hydroxide ions into solution and contribute to neutralize acidity (Chapter 2). This capacity will influence the time development of acidification and should
be included in the sensitivity classification. Those models used to estimate critical loads which include SO$_4^{2-}$ adsorption (MAGIC, SMART, RESAM) commonly assume a molar H$^+$ to SO$_4^{2-}$ adsorption ratio constant at 2.0 (Fumoto & Sverdrup, 2000). However, this ratio is generally less than 2.0, and the resulting net negative charge may affect the base cation dynamics in the soil (Fumoto & Sverdrup, 2001). For Andosols, Fumoto & Sverdrup (2000) considered for implementation in the SAFE model a ratio of ~1.4. This value is higher than the one deduced in this study (H$^+/\text{SO}_4^{2-} = 0.4$ at pH 4, Chapter 8). Moreover, depending on soil initial pH and acid buffering capacity, the H$^+/\text{SO}_4^{2-}$ adsorption ratio may vary significantly. This was demonstrated in Chapter 8, where similar amounts of SO$_4^{2-}$ sorbed on EU2-Bw and VI2-2BC soil samples generated significantly larger OH$^-$ release in the EU2-Bw. It appears that the ability of soils to sorb H$^+$ or release OH$^-$ upon SO$_4^{2-}$ addition needs to be better characterised.

Another aspect is that some tropical plant species have high requirements for sulphur as a nutrient. This could be incorporated into the assessment using a land use or cover map as a second layer to the soil layer. However, this could only be carried out successfully where there are detailed land-cover data for different tree or crop species at national or subnational scales. There are also differences in tolerance of tropical plant species, many of which can tolerate acidic conditions, or are able to counteract acidification (Johnson & Parnell, 1986). However, even the growth of acid-tolerant species is affected by acidification and therefore the degree to which they are affected needs to be determined. Overall, this pleads for improving the models for calculating critical loads in developing countries, where climate, vegetation and soils largely differ from those that prevail in Europe or North-America.

9.2 Counteracting fumigation in the area of Masaya volcano

Air pollution at Masaya is a long-term recurrent problem. Remedies to reduce the emission rates at the vent, or to lessen the impacts of fumigation on vegetation were proposed in the past (McBirney, 1956; references therein). Proposals to control gas emissions at Masaya included sealing of the vent by dynamiting and bombing, diversion of the plume at high altitude by building a 300-m tall chimney and capture the emissions with pipes. Today, Nicaraguan government seems to have abandoned these utopian ideas. On the contrary, present policy does

*Developed from Delmelle *et al.*, 2002
not consider the Masaya area as a priority since the government has planned to use Masaya volcano as a gigantic self-burning rubbish incinerator (Pantoja, 2003). Fortunately, the Instituto Nicaragüense de Estudios Territoriales (INETER) provided evident convincing ecological and security arguments to give up this project.

Rather than directly controlling the fumes at the vent, spraying of lime at the point of damage was suggested as an alternative method to protect vegetation from noxious effects of the acidic gases (McBirney, 1956). A difficulty of these rather-expensive method lies in the necessity to apply frequent sprayings during the wet season. Another solution to preserve coffee and other cultivated plants would be to construct windbreaks of trees and resistant shrubs around the plantation located in the poorly affected areas (Delmelle et al., 2002). Indeed, vegetation act as sink for SO$_2$ and acid aerosol particles (Hill, 1971), windbreaks can also deviate upwards the wind that carries acid gas. This require the use of species that are tolerant to chronic exposure to SO$_2$. The use of Eugenia jambos (McBirney, 1956), Brosimum utile and Clusia rosea (Delmelle et al., 2002) may show to be useful. However, there remains the risk of acute fumigation events, which generate damaging gas concentration to any species. No tests were carried out in the field, but naturally protected areas proved to be less affected by fumigation and are used by farmers.

Although the soil pH range for the growth of cashew (Anacardium occidentale), citrus (Citrus limon) and mango (Mangifera indica) is optimal at 4.5-4.7, these trees can not efficiently replace coffee because of their size that make windbreaks inefficient. Farmers in areas strongly affected by Masaya’s plume are rather making attempt to replace the coffee plants by smaller plants such as pineapple (Ananas bracteatus) and pitahya (Acanthocereus tetragonus) a fruit-bearing cactus (Delmelle et al., 2002). While pitahya plants seems to tolerate the high volcanic air pollutant levels, the harvest of its fruit depends strongly on the blooming period and, thus, on the extent of damage inflicted to flowers by the volcanic emissions (Delmelle et al., 2002). Other plants that could be used include Cassava (Manihot esculenta) and Macadamia (Macadamia tetraphylla).

9.3 Potential environmental impacts of volcanoes

Environmental deterioration caused by volcanic acid gases has been reported in several parts of the world (Chapter 2). Indeed, strong volcanic
sources of SO$_2$, HCl and HF are active on the earth’s surface as shown in Table 9.1, and are likely to generate detrimental effects upon the local, regional and global environment.

Some volcanoes exert a severe impact at the local scale (such as Masaya), other exert a lower influence on the surrounding environment despite higher SO$_2$ emission rates because of their localisation (i.e. Erebus) or their altitude (i.e. Etna). Nevertheless, the impacts of several active or potentially active volcanoes remain poorly documented. Therefore, several detailed and serious studies should be engaged to assess their environmental effects.

In my opinion, two cases deserve particular attention: (i) the possibility that an eruption similar to the 1783 Laki fissure eruption occurs today, and (ii) the poorly documented effects of Nyiragongo and Nyamuragira volcanoes (Democratic Republic of Congo) on the local ecosystems. Based on the results acquired in this PhD, on recent studies and on general knowledge, I have identified several issues of particular interest and drawn elements which may contribute to mitigate some general false ideas.

### 9.3.1 Impacts of the 1783 Laki fissure eruption

In June 1783, a major Icelandic fissure eruption began at Laki volcano. Over 5 to 8 months, an estimated 15 km of basaltic magma and about 61 Tg (S) were emitted as SO$_2$, with about 60% of this released during the first six weeks of the eruption (Stevenson et al., 2003; Thodarson & Self, 2003). The eruption was one of the largest individual tropospheric pollution events of the last 250 years, and the quantity of SO$_2$ released was comparable to the total annual present-day anthropogenic input into the atmosphere (Stevenson et al., 2003). About 60% of the Icelandic grazing livestock died mainly from chronic fluorosis due to the ingestion of F-rich ash that fell in this area. Together with the failure of the harvest, this has led to a famine lasting from 1783 to 1786 that caused the death of ~20% of the Icelandic population. The experience of Iceland in 1783 serves to illustrate how powerful the impact of volcanic gas and aerosol emissions can be in areas relatively close to the volcanic source.

Many sources reported a dry fog or haze over most of Europe, North America and Asia during the second half of the year (Thodarson & Self, 2003; and references therein). The haze had a distinct sulphuric odour, it damaged trees and crops resulting in the failure of the harvest in several regions across Scandinavia and Europe. Moreover, the Laki eruption had a strong climatic effect and reduce surface temperature across Europe and North America by ~1.3 °C for 2-3 years (Thodarson

<table>
<thead>
<tr>
<th>Name of volcano</th>
<th>Activity*</th>
<th>SO$_2$ Flux /t d$^{-1}$</th>
<th>Eruption time /days</th>
<th>Country</th>
</tr>
</thead>
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<tr>
<td>Laki*</td>
<td>explosive</td>
<td>406000</td>
<td>150</td>
<td>Iceland</td>
</tr>
<tr>
<td>Nyamuragira</td>
<td>explosive</td>
<td>244000</td>
<td>808</td>
<td>Dem. Rep. of Congo</td>
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<tr>
<td>Rabaul</td>
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<td>26000</td>
<td>264</td>
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<tr>
<td>Erebus</td>
<td>explosive</td>
<td>25600</td>
<td>5110</td>
<td>Antartica</td>
</tr>
<tr>
<td>Nyiragongo</td>
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<td>23000</td>
<td>156</td>
<td>Dem. Rep. of Congo</td>
</tr>
<tr>
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<td>1229</td>
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<td>5975</td>
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<td><strong>2438</strong></td>
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<tr>
<td>Yasur</td>
<td>explosive</td>
<td>900</td>
<td>5690</td>
<td>Vanuatu</td>
</tr>
</tbody>
</table>

*explosive activity (explosive); passive degassing (passive)
9.3. Potential impacts

The huge area affected by the dry fog illustrates that gases emitted by fissure eruptions can be transported for great distances through the atmosphere and retain sufficient concentration to have a severe environmental impact (Grattan, 2005).

The considerable impacts on crops of Laki eruption is due both to its magnitude and the period when it occurred: June, since it is a critical period for crop growth in the Northern hemisphere. The impacts would have been probably reduced if the eruption had occurred during winter. Today, population (and especially in Iceland) are less dependent on local agriculture and breeding than in the 18th century. The impact that a Laki-type eruption would produce on European or North American agriculture would certainly be lower today than in 1783. However, if a similar volcanic event was to occur in less-developed regions, the impacts would certainly be catastrophic. Indeed, in these areas a large majority of the population depends totally on agriculture. Moreover, in the tropics, agriculture is less seasonal dependant and may last throughout the year. Harvest failure and livestock deaths induced by such eruption would undoubtedly result in a severe famine.

Apparently, the main cause of difficult harvest in 1783 in Europe was the fumigation by the acid haze (Thodarson & Self, 2003). To our knowledge, there was no report of difficult harvest during the season that followed the Laki eruption, suggesting that the soils were not irredeemably affected. According to the model of Stevenson et al. (2003), S deposition over central Greenland was 200 mg S m\(^{-2}\) month\(^{-1}\) in July following the eruption, with a total annual deposition in this area of 360-500 mg S m\(^{-2}\). However, deposition rates rapidly decline to normal levels once the eruption ended. The estimated deposition rates over Greenland and Iceland were particularly large compared to normal levels before the Laki eruption. They are also well above current estimates. However, the deposition rates over Europe and Northern America generated by the Laki fissure eruption can be compared with those due to anthropogenic emissions in the 1990’s.

In order to assess the impact that a Laki scenario would have on European soils, I carried out simulations using the Very Simple Dynamic (VSD) model (Posch et al., 2003). In these calculations, a Belgian Cambisol (Brahy et al., 2000) and a Swedish Podzol (Zysset & Berggren, 2001) were submitted to acidification for a period running from 1800 to 2000 (for simplification, the Laki fissure eruption was considered to occur in 1823). S deposition rates were assumed from Stevenson et al. (2003) and the assumption was made that N depositions were negligible. The simulation results (Figures 9.1 and 9.2) suggest that soils in continental
Chapter 9. Impacts of Volcanic Emissions

Europe were not significantly affected by the acid deposition generated by the Laki emission, compared to today depositions. Even in Scandinavia where intense S deposition probably occurred, impacts on pH and Al/(exchangeable Ca+Mg+K) is similar to that of present-day industrial acidification. These results suggest that, in disagreement with previous studies (e.g. Grattan, 2005), the impacts on European soils were probably limited and occurred only during a short period following the eruption. However, the significant deposition of HCl and HF over Europe generated by the Laki fissure eruption, for which no data are available, should also be included in the simulation.

Undoubtedly, were a Laki fissure eruption to occur today the implications for European environment would be important. However, superimposed to today deposition, this would probably not generate significantly higher acid loading and, to my opinion, not be as catastrophic as predicted by Grattan (2005). This author claimed that an event of this magnitude could conceivably deposit sulphur in excess of the critical threshold for acid loading of soil across Europe. He is right, but depositions are already in excess in Europe, and the additional volcanogenic deposition would not make the difference.

Overall, in Europe, where ecosystems are overstressed and weakened by anthropogenic pollution, the response to an event of the Laki eruption magnitude remains poorly known. Global agriculture is considered vulnerable to sudden global cooling (Engvild, 2003) and would be severely affected by acid fumigation. However, long-term effects on soils and vegetation seem unlikely. Further modelling on soil and ecosystems response to such an event are needed.

9.3.2 Impacts of Nyamuragira and Nyiragongo volcanoes

Recent eruptions of Nyamuragira and Nyiragongo volcanoes have released prodigious amount of SO$_2$ into the atmosphere (Table 9.1; Figure 9.3). On 19 October 1998 and 6 February 2001, Carn & Bluth (2003) observed peak SO$_2$ fluxes from Nyamuragira of 0.2 and 0.77 Tg d$^{-1}$, respectively. These volcanoes are both located in the Virunga National Park (Democratic Republic of Congo), close to areas densely inhabited (300 inhabitants km$^{-2}$). According to an UN report, emissions of gas, ash and cinders from Nyiragongo and Nyamuragira are causing health problems for an estimated 60 000 people in the mountains’ immediate vicinity and about 30 000 km$^2$ of land west of the volcanoes has been destroyed by the fallout. Acid gas emissions have also affected the surrounding environment since in 2001 and 2002 agricultural production decreased by an estimated 60% and some 5000 km$^2$ in the Virunga
9.3. Potential impacts

Figure 9.1: VSD simulation from 1800 to 2000 for a Podzol in Sweden. For simplification, the Laki fissure eruption was considered to occur in 1823.

Figure 9.2: VSD simulation from 1800 to 2000 for a Cambisol in Belgium. For simplification, the Laki fissure eruption was considered to occur in 1823.
National Park were destroyed (IRIN, 2003). The Global Volcanism Program (2003) also reported acid rain in the area with F concentration as high as 15 ppm, 10 to 20 times higher that those reported in Masaya (Annexe 2). Despite the 500 000 people living close to Nyiragongo, no intensive study has been engaged and no concrete initiative has been established to reduce volcanic risks.

![Figure 9.3: Major volcanoes of the Democratic Republic of Congo.](image)

Virunga National Park (covering an area of 790 000 ha) comprises an outstanding diversity of habitats, ranging from swamps and steppes to the snowfields of Rwenzori at an altitude of over 5000 m, and from lava plains to the savannahs on the slopes of volcanoes. Mountain gorillas are found in the park, some 20 000 hippopotamuses live in the rivers and birds from Siberia spend the winter there. This park is part of Africa’s remaining rainforest that contain 10 000 known plant species of which 3000 are endemic (UNDP, 1994). Virunga National Park was inscribed on the List of World Heritage in Danger at the 18th Session of the World Heritage Committee in 1994 in the wake of the war in neighbouring Rwanda and the subsequent massive influx of refugees from that country which led to massive deforestation and poaching at the site. Weakened by the consequences of the war, this ecosystem might be severely affected by the acid volcanic plumes, which will contribute to the decrease in biodiversity observed in this area.

Soils in the vicinity of the volcano are mainly Andosols, but further away (about 20 km distant from the emission source) Ferralsols mantle the area. The chemical fertility of Ferralsols is poor, weatherable minerals are absent and cation retention by the mineral fraction is weak, therefore these soils present lower buffering capacity than Andosols, and are comparatively more sensitive to acid loadings. This would result in progressive Al saturation of the exchange complex, although adsorption
of $\text{SO}_4^{2-}$ could reduce net acidification (Stumpe & Vlek, 1991). Saline and alkaline soils are also found in the Virunga volcanic chain (Van Gysel & Andre, 2004). These soils support a typical vegetation, which may also be severely impacted. However, acid neutralisation should be efficient in these soils. Obviously the extent to which the soils will be impacted depends on acid loading, a largely unknown variable for this area.

As a consequence of the vegetation destruction we might expect an increase in erosion in this area. The loss of plants, which anchor the soil with their roots, may indeed cause widespread erosion. This may results in the decline of crops yields or forest. In turn, people might be tempted to clear additional forest for woods or agriculture and the dramatic erosion cycle could begin.

Virunga National Park was created to protect Primates, which generally engage in geophagy, or soil ingestion. Geophagy is used to alleviating gastrointestinal disorders (Krishnamani & Mahaney, 2000) and as an important source of mineral nutrients, especially iron. Therefore, soils affected by strong F and acid loading could significantly affect the health of primates.

The soils consumed by primates generally have a long weathering history (Krishnamani & Mahaney, 2000), their clay fraction is dominated by kaolin minerals, interstratified kaolinite/smectite, smectitic minerals and iron oxide and aluminous oxide minerals (Wilson, 2003). In the Virunga National Park, gorillas engage occasionally in geophagy (Krishnamani & Mahaney, 2000) and use volcanic soils (Schaller, 1963). Gorillas are found throughout the six dormant volcanoes of the Virunga National Park, but not on the two active ones (Schaller, 1963; Kalpers et al., 2003). Depending on wind direction, the territory where gorilla live may be affected by the gas plume emitted by Nyiragongo and Nyamuragira volcanoes.

According to Simon (1998) soil ingestion by human varies between 1 to 300 g day$^{-1}$. It can be reasonably assumed that about 200 g of soil are ingested daily by an adult gorilla. Based on the account of Schaller (1963), the soils ingested by gorillas comprise a significant fraction of Andosols. This soil material may contain significant amounts of fluoride; for example 50 µmol oxalate-extractable F per g of soil were measured in the Masaya Eutric Andosols not exposed to volcanic deposition (Chapter 4). Therefore, gorilla in the Virunga National Park may ingest about 190 mg F per day (or about 0.95 mg F kg$^{-1}$ body-weight day$^{-1}$). This is by far larger than the human Reference Dose for Chronic Oral Exposure for Fluoride (RfD; 0.06 mg kg$^{-1}$ day$^{-1}$), which is an estimate of a daily exposure to the human population that is likely
to be without an appreciable risk of deleterious effects during a lifetime (IRIS, 2004). Exposure to such a dose of fluoride is potentially harmful for the health of the gorilla. It may give rise to dental and skeletal fluorosis. Skeletal fluorosis is a health effect of excessive accumulation of fluoride in bones leading to changes in bone structure and making them extremely weak and brittle. The development of crippling skeletal fluorosis in human requires the consumption of 0.28 mg kg$^{-1}$ day$^{-1}$ of fluoride over a 20-year period (U.S. EPA, 1985).

Any threat of environmental degradation to such an area must, therefore, be closely monitored. As many of these areas are not only inaccessible but also dangerous for observers to visit, an alternative option is the use of satellite imagery to report regions of degradation.

Overall, these two examples call for detailed and serious studies to assess the actual and potential environmental, agronomical and economical effects of volcanic degassing worldwide.