Category		Title
NFR:	**11.A**	**Volcanoes**
SNAP:	**1108** **110800**	**Volcanoes** **Volcanoes**
ISIC:		
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Contents

1   Overview

This chapter describes emissions from geothermal activities, both eruptive and non-eruptive. Sources include not only volcanoes, but also fumaroles, geysers, metamorphic degassing or other activities related to molten magma in the earth's crust. Heated magna under pressure contains gases like sulphur dioxide, carbon dioxide, hydrogen sulphide, mercury and chlorine. These gases may be released when magma gets close to the surface and pressure may be discharged.

With respect to the different sources, non-eruptive volcanoes that outgas at relatively constant rates seem to be more important than those from sporadic eruptions, both for CO_2 (Gerlach, 1990) and SO_2 (Andres and Kasgnoc, 1997). However, the sporadic emissions are much more difficult to assess.

Some of the emissions may also be considered anthropogenic when produced at geothermal power plants where artificial holes are drilled to obtain hot water from the earth's interior. These emissions, however, are treated in NFR source category 1.B.2.a.vi, Geothermal energy extraction, and are assumed to be rather small.

Emissions from volcanoes show great regional and temporal variation. Most affected are volcanic areas, and also volcanic activity tends to be highly variable. The number of active subaerial volcanoes per year based on a five-year running average is approximately 60 (Andres and Kasgnoc, 1997; Simkin and Siebert, 1984). Globally, SO_2 emissions from volcanoes are estimated to account for about 10–15 % of the anthropogenic flux (Halmer et al., 2002). Emissions of CO_2 from volcanoes are considered about two orders of magnitude lower than the anthropogenic output of CO_2 (Gerlach, 1990). Considerable emissions of aerosols are present in most volcanic plumes (Ammann et al., 1990). Emissions of Hg, Cl_2 and F_2 have been measured occasionally, but are very difficult to generalize.

2   Description of sources

2.1 Process description

Heated rocks in the earth's crust may be chemically transformed such that gases are released. Carbonates may thus release CO_2, and sulphates SO_2. These gases may be dissolved at a high pressure in the molten magma. Reaching the surface (either at the sea floor for submarine volcanoes, or at the atmosphere) the pressure decreases and the gases are emitted into the atmosphere.

2.2 Techniques

A differentiation of techniques is not applicable to natural emission sources. However, different source categories exist. Volcanoes are sources that have magma outflow. By contrast, fumaroles and other sources only vent gases through cracks in the rocks.

There are also significantly different emission patterns among volcanoes. Outgassing may occur continuously (globally the larger portion of emissions), or are episodic in the course of an eruption. Differentiation can also be made among eruptive emissions; eruptions in an arc tectonic

regime tend to be more violent, but seem to have a more predictable pattern of explosivity strength vs. SO_2 emissions.

The different types of volcanoes are well known and data are available. Generally, continuous flow volcanoes have low viscosity magma and therefore also flat slopes, while eruptive volcanoes are comparatively steep.

2.3 Emissions

Volcanoes release considerable fluxes of gases and particles to the atmosphere, both during eruptions and by long-term noneruptive degassing. The most important species released directly from magma at high temperatures are SO_2, H_2O, CO_2; trace constituents include HCl, HF, Hg, CuCl, etc (Etiope and Klusman, 2002). Volcanic emissions also include species produced in the extreme environments associated with the volcano. Observations of NO_x flux associated with volcanic activity suggest that this is the result of thermal oxidation of NO formed via reaction of N_2 (from the atmosphere as well as magmatic sources) and O_2 (from the atmosphere), followed by rapid oxidation of the product NO (Pyle et al., 2005).

Particulate emissions may originate from (Mather et al., 2004):

- pyroclastic material (tephra);

- condensation of volcanic gases, as they cool;

- transformation of existing particles;

- low-temperature reactions (gas-to-particle reactions at ambient temperature and aqueous phase reactions).

Methane emissions from geothermal reservoirs originate through bacterial and thermal decomposition of organic matter, as well as inorganic synthesis (Fischer-Tropsch type: $CO_2 + 4H_2 = CH_4 + 2H_2O$) and outgassing from the mantle (Etiope and Klusman, 2002).

2.4 Controls

There are no controls to natural emissions by definition.

3 Methods

3.1 Choice of method

There is very little information available on emissions from volcanoes and the information available does not allow us to identify a Tier approach as in the other sectoral chapters. The Tier 1 and Tier 2 do not contain emission factors, and emission can be estimated only using the Tier 3 approach.

3.2 Tier 1 default approach

3.2.1 Algorithm

The Tier 1 approach for emissions from this source category uses the general equation:

$$E_{pollutant} = AR_{production} \times EF_{pollutant} \qquad (1)$$

This equation is applied at the national level, using annual national statistics on volcano activity. However, for this source category it is difficult to identify activity rates.

3.2.2 Default emission factors

The main emission from volcanoes is SO_x. However, no Tier 1 estimate is available since there is no simple emission factor available for calculating total SO_x emissions from volcanoes. Therefore, the emission of SO_x is listed as 'Not Estimated' in the table below. As stated in subsection 2.3 of the present chapter, other trace elements may be emitted as well, but no emission factors are available.

To estimate emissions from volcanoes, it is good practice to use the approach described in the Tier 3 section.

Table 3-1   Tier 1 emission factors for source category 11.A Volcanoes

Tier 1 default emission factors		
	Code	Name
NFR Source Category	11.A	Volcanoes (11.08)
Fuel	NA	
Not applicable	Aldrin, Chlordane, Chlordecone, Dieldrin, Endrin, Heptachlor, Heptabromo-biphenyl, Mirex, Toxaphene, HCH, DDT, PCB	
Not estimated	NOx, CO, NMVOC, SOx, NH3, TSP, PM10, PM2.5, Pb, Cd, Hg, As, Cr, Cu, Ni, Se, Zn, PCDD/F, Benzo(a)pyrene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Indeno(1,2,3-cd)pyrene, Total 4 PAHs, HCB, PCP, SCCP	

3.2.3 Activity data

There are no statistical data available. Instead, geological information needs to be obtained from the respective national geological survey.

Satellite data can in principle be obtained from the National Aeronautics and Space Administration (NASA) or the National Oceanic and Atmospheric Administration (NOAA), respectively. The exact procedures however have not been checked.

3.3 Tier 2 technology-specific approach

Tier 2 is not available for this source category. To estimate emissions from volcanoes, it is good practice to use the approach described in the Tier 3 section.

3.4 Tier 3 emission modelling and use of facility data

3.4.1 Methodology description

This section describes the methodology to estimate emissions from volcanic activity. It has been split in two parts: the first part describes the state-of-the-art methodology and the second part discusses an improved methodology.

3.4.1.1 State-of-the-art methodology

Primary source of geothermal emissions are active volcanoes. These volcanoes are well known and geologically described. Great efforts have been made in the development of systematic plume measurements in volcano monitoring programs. In particular, Kilauea and Mount St. Helens have nearly continuous records of SO_2 fluxes since 1979 and 1980 respectively (Malinconico, 1987), and continuous monitoring systems are in place for Mount Etna and Stromboli since 2002 (INGV (Italian National Institute for Geophysics and Volcanology), 2005). SO_2 emissions are usually assessed using spectrometric data (Hoff and Gallant, 1980) from correlation spectrometers (Cospec) (Gerlach and McGee, 1994) obtained by means of ground-based stationary and mobile techniques, or airborne techniques, also in combination with available satellite data (Gerlach and McGee, 1994). Cospec is useful under many field and volcanic conditions, but is used most routinely under quiet to mildly explosive conditions. Cospec is not routinely used in the largest volcanic eruptions due to logistical and instrumental limitations. However, large eruptions can sometimes be monitored by satellite methods, especially with the total ozone mapping spectrometer (TOMS) (Krueger et al., 1995). Like Cospec, TOMS measures SO_2 emissions only. A variety of remote-sensing and direct sampling techniques has been employed in the measurement of volcanic aerosol and the characterisation of volcanic plumes (Mather et al., 2004).

An extensive compilation of available, measured volcanic S fluxes has been carried out for the Global Emissions Inventory Activity (GEIA) (Andres and Kasgnoc, 1997). The data set contains volcanic SO_2 emissions averaged over the 25 years from the early 1970s to 1997, based on Cospec measurements. It includes average SO_2 emissions from 49 continuously emitting volcanoes (four located in Europe: Etna, Stromboli, Vulcano and Kverkfjoll) and maximum SO_2 emissions from 25 sporadically emitting volcanoes (none located in Europe). This information can be extrapolated to provide SO_2 emission figures for the ~ 300 currently active volcanoes. SO_2 emissions from explosive volcanism can be assessed based on the Volcanic Explosivity Index (VEI) of volcanoes. The VEI is based on the height of the eruption column and the volume of material ejected; it is an open-end scale from VEI 0 for small explosive eruption to VEI 8 for the largest known historic eruption (Newhall and Self, 1982).

The Smithsonian Global Volcanism Network catalogues each eruption during the past 200 years and provides a value for the Volcanic Explosivity Index for each singular eruption (www.volcano.si.edu/gvp/). Differentiation is to be made between arc-volcanoes and non-arc volcanoes. It is good practice to scale emissions from emitting volcanoes to one of those listed in the data set (Andres and Kasgnoc, 1997).

The compilation also includes average mass ratios to SO_2 for five sulphur species (H_2S, CS_2, OCS, SO_4^{2-} and particulate S) which can be used to estimate average fluxes. CO_2 emissions may also be derived from SO_2 emissions, considering the additional uncertainties. The secondary sources (fumaroles, geysers) are hardly ever significant sources, except for methane. It is good practice to estimate diffuse emissions on the basis of the average value of the gas flow per surface unit and of the area of land where the phenomenon occurs, whereas emissions from vents can be estimated from approximations of the number of sources, the volume gas flow and the concentrations. Etiope and Klusman (2002) have collected available data, both for diffuse soil degassing and for gas vents.

3.4.1.2 Improved methodology

To estimate SO_2 emissions from explosive volcanism, Schnetzler et al. (1997) have proposed a 'VSI' (Volcanic Sulphur dioxide Index). In contrast to the VEI, the VSI relates directly to the amount of volcanic SO_2 produced. It is scaled to be as compatible as possible with the VEI, and allows for differentiation between arc and non-arc volcanoes.
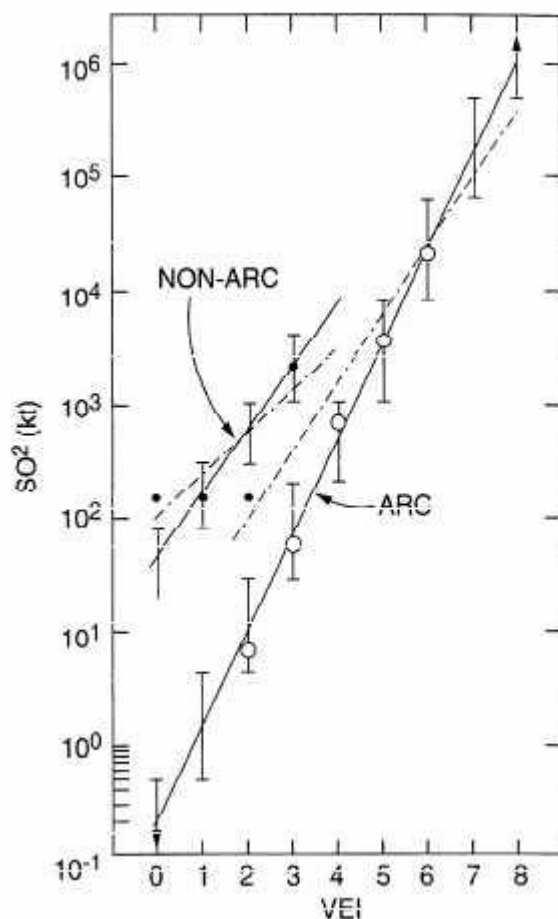

Figure 3-1   Average SO_2 emissions of volcanic eruptions as a function of VEI for arc and non-arc volcanoes (Schnetzler et al., 1997)

Halmer et al. (2002) have compiled a global data set of volcanic degassing during both explosive and quiescent volcanic events. They have modified the original VSI by multiplying it with a factor of approximately 2 to match the values of measured SO_2 emissions. For continuously emitting volcanoes, the assessment has been based on the following parameters referred to monitored volcanoes:

* stage of activity (silent to explosive);
* tectonic setting (subduction zone, rift zone and ocean island);
* magma composition (basaltic to highly differentiated).

The data set also includes a semi quantitative estimate of other gas components emitted, based on SO_2 fluxes and known molar ratios (e.g. H_2S/SO_2), according to the assumption that the different gas components emitted by a volcano are in equilibrium with each other and that the molar ratios

of the gas species in high-temperature fumaroles are similar to molar ratios equilibrated at depth where the gas separates from the magma.

3.4.2   Tier 3 emission calculation

For explosive emissions, the following relationship has been developed (Bluth et al., 1993):

$$\log E = -0.25 + 0.76 \ VEI \tag{2}$$

where:

- E is emission amount of SO_2 (kt),
- VEI is the volcanic explosivity index.

It only applies to arc volcanoes. For non arc-volcanoes, emitted SO_2 is typically much higher and less dependent on the VEI. It is good practice to assume an order of magnitude higher emission for eruptions of non-arc volcanoes, using the same formula as for arc volcanoes. The uncertainty is very high, however. As an alternative, the VSI concept can be used, on the basis of Table 3-2 (Schnetzler et al., 1997).

Table 3-2   Volcanic SO_2 index (VSI) as proposed by Schnetzler et al. (1997) compared with the VEI scale of Newhall and Self (1982)

VSI	0	1	2	3	4	5	6	7	8
Arc volcano SO_2 [kt]	< 0.5	0.5–4	4–30	30–200	200–1 000	1 000–8 000	8–60 10^3	60–500 $\cdot 10^3$	> 500 $\cdot 10^3$
Non-arc volcano SO_2 [kt]	< 80	80–300	300–1000	1 000–4 000					
VEI	**0**	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**
General descr.	Non-explosive	Small	Moderate	Moderate large	Large	Very large	Very large	Very large	Very large
Cloud column height [km]	< 0.1	0.1–1	1–5	3–15	10–25	> 25	> 25	> 25	> 25
Volume of tephra [m^3] (arc only)	< 10^4	10^4–10^6	10^6–10^7	10^7–10^8	10^8–10^9	10^9–10^{10}	10^{10}–10^{11}	10^{11}–10^{12}	10^{12}–10^{13}

The modified VSI index proposed by Halmer et al. (2002) is shown in Table 3-3; the original VSI values have been multiplied with a factor of approximately 2, to match the values of measured SO_2 emissions.

Table 3-3   Modified VSI as proposed by Halmer et al. (2002)

VSI	0	1	2	3	4	5	6	7	8
Subduction zone volcanoes SO_2 [kt]	< 1	1–8	8–60	60–800	200–2 000	1–16 $\cdot 10^3$	16–120 $\cdot 10^3$	120–$1\,000$ $\cdot 10^3$	$> 1\,000$ $\cdot 10^3$
Other volcanoes SO_2 [kt]	< 160	160–600	600–2 000	2 000–8 000					

For non-explosive emissions, it is good practice to estimate SO_2 emissions on the basis of the information available for monitored volcanoes, taking into account the following parameters: stage of activity, tectonic setting, and magma composition (Halmer et al., 2002).

CO_2 emissions are in the range of 0.01–0.05 10^{12} mol/yr (0.44–2.2 Mt/yr) for one major volcano (Gerlach, 1991), but occasionally much higher (Mt. Etna: plume emissions and diffusive emissions combined 25 Mt/yr). The ratio of CO_2/SO_2 is considered to be around 1.5 for arc emissions, but globally up to 4 or 5 on the molar scale (Gerlach, 1991), part of the difference caused by the unusually high CO_2 emissions from Mt. Etna. These figures may be taken if no other information is available.

According to Andres and Kasgnoc (1997), the ratio of SO_2-S to S in other sulphur species is about 2:1, with 71 % of the sulphur contained in H_2S. The mass ratio of H_2S/SO_2 is 0.21 and may be applied for estimating H_2S emissions. Mass ratios for other S compounds rely on only a few measurements and therefore are prone to many error sources.

SO_2/HCl ratios in volcanic fluids (either fumaroles or plumes) are often studied, since they can provide precious insights into volcanic processes, e.g. as concerns magma composition and the interaction of magmatic gases with hydrothermal systems. Plume measurements available for Mount Etna between 1992 and 1995, and in 2002–2003, both for eruptive and non-eruptive activity periods, show fairly constant HCl/HF ratios, but wide variation in the SO_2/HCl ratios (Pennisi and Le Cloarec, 1998; Aiuppa et al., 2004). Samples collected between 1992 and 1995 are characterised by HCl/HF ratios between 5 and 14 (Pennisi and Le Cloarec, 1998). During May–June 2002, the SO_2/HCl molar ratio in plume measurements for Mount Etna consistently showed a value of 4.5±1, whereas the HCl/HF molar ratio varied in the range from 6 to 12 (Aiuppa et al., 2004). During October 2002 to February 2003 eruption of Mount Etna, the SO_2/HCl molar ratio suddenly increased from 4 to 8 in December 2002, and then decreased throughout the eruptive period to 1.5 in February 2003. It is good practice to consider the rather constant HCl/HF ratio observed throughout the period as a characteristic feature of the volcano for further applications (Pennisi and Le Cloarec, 1998). An average SO_2/HF plume mass ratio of about 27 has been reported (Pennisi and Le Cloarec, 1998; Aiuppa et al., 2004; Francis et al., 1998; Aiuppa et al., 2001; Burton et al., 2003).

F and Cl emission data are available for Mt. Erebus, Antarctica, which is has a very uncommon alkaline magma, rich in halogens and various trace metals. These data therefore need to be seen as an indication of an upper boundary rather than as an emission factor as such. The average F/S ratio (by weight) in Erebus gas reported is 0.69, for Cl/S it is 0.55 (Zreda-Gostynska et al., 1993). Similarly, Hekla (Iceland) is renowned for its high concentration of F and Cl during eruption. In the compilation of their data set, Halmer et al. (2002) have used the molar ratios listed in Table 3-4 (however, it is good practice to use these results only if no specific information is available).

Table 3-4   Molar ratios for halogen compounds released from volcanoes (Halmer et al., 2002)

	SO_2 / HCl	SO_2 / HF	HBr / HCl
Subduction zone-related volcanoes	0.1–10	2–18	0.4–2.4 $\cdot 10^{-3}$
Other volcanoes	97	70	

Recent HNO_3 measurements in volcano plumes have provided mean molar HNO_3/SO_2 ratios of 0.01, 0.02, 0.05 and 0.07 for Villarrica, Masaya, Etna and Lascar respectively (Mather et al., 2004). Emissions of heavy metals from volcanoes are generally estimated on the basis of the metal/sulphur ratio; however, available measurements cannot be generalised. Emissions of 4–20 mg Hg/kg fumarole vapours have been reported and may be applied (Ferrara et al., 1994).

Particle flux estimates for individual volcanoes reveal orders of magnitude of variation, even for the same volcano in the same phase of activity (Mather et al., 2003). Measurements performed near the summit of Mount Etna in October 1997 using an eight channel sun-tracking photometer indicate a trimodal distribution consisting of particles of radius < 0.1 μm (nucleation mode), a possible second fine mode (< 1.0 μm), and large particles of radius > 1.0 μm (acid droplets) with minima at 0.5 and 1.5 μm. The mean effective radius was determined to be at 0.83 μm within the range 0.35 < r < 1.6 μm, and the total aerosol mass flux was estimated as 4.61–7.91 kg/s with the smaller radius mode contributing 6–18 % of the total mass flux; the corresponding estimate for sulphate flux was 0.5–0.8 kg/s, with the SO_2 flux amounting to around 56.4 kg/s (Watson and Oppenheimer, 2001).

CO_2 emissions from geothermal fields have been reported in Haraden (1989). Strictly speaking, these emissions are anthropogenic and it is good practice to report them in connection with power generation (Chapter 1.A.1, Energy industries).

3.4.3   Activity data

The Smithsonian Global Volcanism Network (www.volcano.si.edu/) provides information for each volcanic eruption during the past 200 years, as well as dates and explosive magnitudes of large eruptions during the Holocene (the last 10 000 years). A value for the Volcanic Explosivity Index is reported for each singular eruption. For each volcano, descriptions of the volcanic activity are also available, through monthly and weekly reports. As concerns Italian volcanoes, SO_2 fluxes from Mount Etna have been available since 1975. For the period 1975–1987, they were based on episodic Cospec measurements reported by three different sources (Allard et al., 1991; Caltabiano et al., 1994; Allard, 1997). Since1988, regular measurements (four measurements every month) have been carried out by the National Institute of Geophysics and Volcanology (INGV) ——— Catania Section (Bruno et al., 1999).

4 Data quality

4.1 Completeness

No specific issues.

4.2 Avoiding double counting with other sectors

No specific issues.

4.3 Verification

Petrologic estimates of volatile emissions during eruptions, based on the volatile content of melt inclusions trapped in crystals, may be used to verify data obtained from spectroscopic measurements (Gerlach et al., 1994). However, this approach generally provides minimum estimates.

4.4 Developing a consistent time series and recalculation

No generalization possible; for instance, some volcanoes, such as Mt. Etna, have a memory of their previous behaviour, whereas other display totally random behaviour, with no correlation between repose time and eruption size. Temporal disaggregation may be performed for past periods according to available records of volcanic activity.

4.5 Uncertainty assessment

The measured variability of diffuse emission fluxes may be in the order of 20 % (relative standard deviation, Allard et al. 1991). In addition to this, it is good practice to take into account that the total uncertainty of an individual Cospec measurement, not including signal attenuation, generally ranges from ±15 to ±25 %, and in the worst case observed has been approximately ±25 % (Malinconico, 1987). The uncertainty with the emission factors, however, is assumed to be in the range of one order of magnitude. Ratios of SO_2-S to S in the other sulphur compounds and to other trace species rely on only a few measurements and therefore no quantitative estimation of uncertainty can be provided.

4.6 Inventory quality assurance/quality control QA/QC

Surrogate parameters to establish emission factors are rather weak. Validation of Cospec measurements through TOMS data and petrologic estimates for assessing eruptive emissions is needed. Available measurements do not allow generalization of ratios to SO_2-S for other S compounds and other trace species. This applies, in particular, to metal/sulphur ratios, which vary not only from volcano to volcano, but also temporally and spatially at any site, thus showing decoupling of the metal-rich particulate phase from the S-rich gas phase. Volcanic plume geochemical studies in the past have tended to focus on the measurement of composition and emission rates of gases. In order to improve our understanding of volcanic particles emissions, future field experiments should aim at characterizing the size-resolved chemistry of aerosol, gas particle interactions between plume constituents and the ambient atmosphere (including rural and urban atmospheres, and transport and deposition of both gaseous and particulate volcanogenic components) (Mather et al., 2004).

Table 4-1   Uncertainty parameters

Parameter	Best estimate	High	Low	Quality
SO_2 measured emissions from Etna [kt/yr]	1 779	3 741	428	B
SO_2 measured emissions from Stromboli [kt/yr]	110	113	107	B
SO_2 measured emissions from Vulcano [kt/yr]	6.9	9.5	5.5	B
Annual average SO_2 estimated emissions from Icelandic volcanoes* [kt/yr]	1 225	1 962	488	D
Mass ratio of H_2S-S to SO_2	0.21	0.50	0.0024	C
Mass ratio of SO_4^{2}-S to SO_2	0.034	0.060	0.0058	C
Mass ratio of particulate S to SO_2	0.0060	0.010	0.0026	C
Mass ratio of CS_2-S to SO_2	0.022	0.022	0.022	D
Mass ratio of OCS-S to SO_2	0.022	0.022	0.022	D
Molar ratio of SO_2 to HCl for Etna	5.05	10	0.1	C
Molar ratio of SO_2 to HCl for Icelandic volcanoes	97	97	97	C
Molar ratio of HCl to HF for Etna	9.25	14	5	C
Molar ratio of SO_2 to HF for Icelandic volcanoes	70	70	70	C
Molar ratio of HCl to HBr for Etna	$1.4 \cdot 10^{-3}$	$2.4 \cdot 10^{-3}$	$0.4 \cdot 10^{-3}$	C
Molar ratio of HCl to HBr for Icelandic volcanoes	$1.4 \cdot 10^{-3}$	$2.4 \cdot 10^{-3}$	$0.4 \cdot 10^{-3}$	C
Molar ratio of HNO_3 to SO_2 for Etna	0.05	0.14	0.00	D
Molar ratio of total particles to SO_2 for Etna	0.1110	0.1402	0.0817	D

Note:
* Using the modified VSI as proposed by Halmer et al. (2002).

4.7 Gridding

It is good practice to limit emission areas to geologically active areas, like calderas.

4.8 Reporting and documentation

No specific issues.

5 Glossary

Volcano	Site where molten magma/lava occasionally reaches the surface.
Non-arc volcano	Volcano on a hot spot or rift zone — erupts more frequently, total number is smaller.
Arc volcano	Volcano at a subduction zone — eruptions are more violent.
Fumarole	Gas vent caused by leaks from magma underneath.
Geyser	Water fountain driven by venting gas due to hot magma.

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7   Point of enquiry

Enquiries concerning this chapter should be directed to the relevant leader(s) of the Task Force on Emission Inventories and Projection's expert panel on Agriculture and Nature. Please refer to the TFEIP website (www.tfeip-secretariat.org/) for the contact details of the current expert panel leaders.