Ozone depletion in tropospheric volcanic plumes

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[1]   We measured ozone (O_3) concentrations in the atmospheric plumes of the volcanoes St. Augustine (1976), Mt. Etna (2004, 2009) and Eyjafjallajökull (2010) and found O_3 to be strongly depleted compared to the background at each volcano. At Mt. Etna O_3 was depleted within tens of seconds from the crater, the age of the St. Augustine plumes was on the order of hours, whereas the O_3 destruction in the plume of Eyjafjallajökull was maintained in 1–9 day old plumes. The most likely cause for this O_3 destruction are catalytic bromine reactions as suggested by a model that manages to reproduce the very early destruction of O_3 but also shows that O_3 destruction is ongoing for several days. Given the observed rapid and sustained destruction of O_3, heterogeneous loss of O_3 on ash is unlikely to be important. **Citation:** Vance, A., A. J. S. McGonigle, A. Aiuppa, J. L. Stith, K. Turnbull, and R. von Glasow (2010), Ozone depletion in tropospheric volcanic plumes, *Geophys. Res. Lett.*, *37*, LXXXXX, doi:10.1029/2010GL044997.

1.   Introduction

[2]   Volcanoes have long been recognised as major sources for gases and particles for the atmosphere. However for many years little attention was paid to their impact on the oxidation capacity of the troposphere. *Bobrowski et al.* [2003] showed the presence of very large amounts of bromine oxide (BrO) radicals in the plume of Soufrière Hills, Montserrat during quiescent degassing. Several other studies found BrO in the plumes of other passively degassing volcanoes [e.g., *Oppenheimer et al.*, 2006; *Bobrowski et al.*, 2007] and satellite observation also confirmed the presence of BrO in the plume of Kasatochi volcano after its explosive eruption in 2008 [*Theys et al.*, 2009]. From other parts of the atmosphere we know that BrO mixing ratios of 1–10% of the values in volcanic plumes lead to very strong catalytic destruction of ozone (O_3) [see, e.g., *von Glasow and Crutzen*, 2007], hence one would expect O_3 depletion also to occur in volcanic plumes.

[3]   Very little data is available about O_3 in volcanic plumes mainly due to logistical difficulties of access and cross-sensitivities of many O_3 instruments to SO_2: Broadband instruments measuring integrated UV absorption may have a positive interference with SO_2, whereas wet-chemical methods as often used in balloon soundings have a negative cross-sensitivity on a mole-per-mole basis [*Schenkel and Broder*, 1982]. *Hobbs et al.* [1982] used an interference-free chemiluminescence technique and reported a 90% depletion of O_3 in the plume of Mt. St. Helens compared to the background air, however they give very little details on this. *Zerefos et al.* [2006] found a strong O_3 depletion in Mt. Etna's plume over Greece, however they used a wet-chemical technique so the lack of O_3 in their data might simply imply the presence of a volcanic SO_2 layer. O_3 depletion in stratospheric volcanic plumes has also been observed [e.g., *Rose et al.*, 2006].

[4]   A number of modelling studies [*Bobrowski et al.*, 2007; *Roberts et al.*, 2009; *von Glasow*, 2010] has aimed at reproducing the observed halogen radicals in volcanic plumes and predict significant O_3 destruction in the plume. The model used by *von Glasow* [2010] was initialised mainly based on observations at Mt. Etna. The results showed very strong O_3 destruction in volcanic plumes for the whole duration of the model runs (3 days). More details about reactive chemistry in volcanic plumes can be found in the recent review by *von Glasow et al.* [2009]. In this paper we present data from a variety of instruments, volcanic settings and different campaigns that clearly show a strong O_3 depletion in volcanic plumes.

2.   Field Sites and Methods

2.1.   St. Augustine (February 1976)

[5]   Airborne in situ O_3 data were collected by the University of Washington (UW) Cloud and Aerosol research group during the January–February 1976 eruption of the St. Augustine Volcano, located in the lower Cook Inlet, southwest of Anchorage, Alaska.

[6]   Flight tracks were generally arranged as a series of cross plume traverses at different altitudes, designed to map out the plume vertical extent, or repeated passes through 'puffs' as they moved downwind. More information on the B-23 sampling and the 1976 St. Augustine eruption is provided by *Hobbs et al.* [1977] and *Stith et al.* [1978, and references therein].

[7]   Ozone measurements were made with a commercial chemiluminescence O_3 analyzer (Monitor Labs model 8410A), which measures O_3 by sensing light output from the reaction of O_3 with ethylene. This technique has no known interferences with volcanic gasses but might over-read O_3 by a few % at high humidities, so that our reported O_3 losses are lower limits. It had a response time of less than 5s for 0 to 200 ppbv. The analyzer was calibrated with an UV-lamp-based calibrator (see *Hegg et al.* [1976] for more details).

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t1.1 **Table 1.** Airborne Measurements of the O$_3$ Depletion in the Plume
t1.2 of St. Augustine, 1976[a]

Date	O$_3$ Loss	State	Distance	Alt	Part. Flux
09 Feb	20–50%	BE			400
11 Feb	67–87%	BE			200
12 Feb	0%	BE			2
13 Feb	44–89%	BE, ash	~24km	2.1–2.4km	60–3 × 10^5
14 Feb	78–100%	PE	28–56km	1.8–2.4km	30 – 90
16 Feb	52%	PE			60
18 Feb	23–67%	PE	~56km	2.1–2.5	20–30

t1.11 [a]"O$_3$ loss" is relative depletion of O$_3$ compared to background measure-
t1.12 ments, "Distance" is from the crater and "Alt" is the altitude above sea level.
t1.13 "State" is the eruption state: ME - major eruption, BE - between eruptions,
t1.14 PE - mostly post eruptive, ash - high ash loading and "Part. Flux" is the esti-
t1.15 mated flux (or range of fluxes, when more than one measurement was made)
t1.16 of particulate matter (in kg s^{-1}) measured on a given day after *Stith et al.*
t1.17 [1978].

2.2. Etna (2004, 2009)

[8] Mt. Etna, one of the largest volcanic gas point sources on Earth, was selected as archetype of open-vent persistently active basaltic volcanoes. The results reported here were acquired over two field campaigns in July–August 2004 and July 2009. Our measurements were performed during phases of passive (quiescent) degassing activity from the volcano's summit vents: The 2004 campaign was performed a few months prior to an effusive eruption affecting the volcano's eastern flank, from 09 Sept 2004 to 08 March 2005. The 2009 campaign was carried out only a few weeks after the end of the most recent eruption (the May 2008–July 2009 effusive event). Etna's average gas output during our observations was generally at (or somewhat below) the ordinary degassing style of the volcano and the Central and North East craters were the most actively degassing open vents.

[9] In the 2004 campaign, we deployed 19 diffusion tubes with the objective of determining atmospheric O$_3$ concentrations near the summit craters as well as on the up- and downwind flanks. Diffusion tubes are passive air sampling devices that rely on the molecular diffusion of the species of interest through an entrapped air volume (for more details see the auxiliary material and *Aiuppa et al.* [2007]).[1]

[10] In 2009 we made O$_3$ measurements at the North East Crater and on the flanks of the South East and Central Craters which were at that point in the plumes of the North East and the Central Crater. We used an UV absorption instrument (2B Technologies, Boulder, CO, USA, model 202) with CrO$_3$ scrubbers for SO$_2$ (EnSci, Boulder, CO, USA) as this instrument shows a strong positive cross-sensitivity to SO$_2$ (at the very high SO$_2$ concentrations near the craters) which absorbs in the same wavelength range. The uncertainty of the 2B instrument is about ± 2 ppbv. Additionally measurements of the aerosol size distribution were made with an optical particle counter (TSI, AeroTrac 8220) with a size range of d = 0.3−10.0 μm. The sampling efficiency at d = 0.3 μm is reported as 50% and 100% at d = 0.45 μm.

2.3. Eyjafjallajökull (April–May 2010)

[11] The data from Eyjafjallajökull were collected between 20 April and 18 May 2010, when the BAe 146-301

operated by the Facility for Airborne Atmospheric Measurements (FAAM) made nine flights involving 45 transects of the Eyjafjallajökull plume, aged between 1 and 9 days old, as it was advected over the United Kingdom and northern France. The plume was generated via eruptive activity at Eyjafjallajökull, which commenced on 20 March. Lava extrusion through fissures resulted in Hawaiian style fire fountains, and generation of phreatomagmatic ash and gas plumes which rose up to 8 km. This event is publicly memorable for having resulted in an unprecedented closure of much of European airspace. Spatially, the plume, intersected during the measurements at altitudes between ~4–8 km, was highly heterogeneous often consisting of multiple layers, suggestive of the recirculation within the persisting high pressure system, as observed by a number of satellite instruments.

[12] SO$_2$ and O$_3$ were measured using Thermo Environmental Instruments Inc. Models 43 and 49C, respectively. The former is based on pulsed fluorescence and the latter on optical absorption. Contamination of the O$_3$ signal by atmospheric SO$_2$ is a known issue with such measurements; however in this case this does not apply as the 49C operates with dual cells: the reference and the sample, with the O$_3$ being chemically scrubbed in the former. The difference between observed absorption in both cases is used to infer the O$_3$ concentration. Therefore any interfering absorption from SO$_2$ is cancelled out. The lowest detectable limit of O$_3$ is 1 ppbv and the overall 2σ uncertainty is estimated at ±3 ppbv. The SO$_2$ sensor is specified as having a lower than detection limit response to ambient ozone levels, ruling out cross-sensitivity in the opposite direction. The overall uncertainty for the SO$_2$ instrument is 7–10%. The design of the sample inlet for the gas analysers is such that no significant aerosol reaches the instruments; this has been confirmed by inspection of the PTFE membrane filter fitted in the sample line immediately upstream. These data were augmented with simultaneously collected nephelometric proxies for plume mineral aerosol loading (from a separate inlet), derived from a three wavelength TSI Inc. 3563 unit. The inlet used with this instrument becomes increasingly inefficient for particles with diameter greater than approximately 5 μm. The largest source of error in data from both Thermo instruments is likely to result from rapid changes in gas concentration as the instruments' integration times are 30 and 50 seconds for O$_3$ and SO$_2$, respectively.

3. Results

3.1. St. Augustine

[13] A summary of the data from 27 plume interceptions is listed in Table 1 (all interceptions are shown in Table S1 in the auxiliary material). The O$_3$ loss relative to the background values is on average 60% but has a very large variability (0–100%), which probably reflects differences in the plume composition on the different days but also different chemical processing due to different atmospheric conditions, e.g., mixing.

3.2. Etna

[14] The diffusion tube measurements at Mt. Etna in 2004 (Figure 1) show very distinct loss of O$_3$ in the first 3.5–4 km from the summit craters. Very often, except for conditions with very low wind speeds, the plume from Mt. Etna's
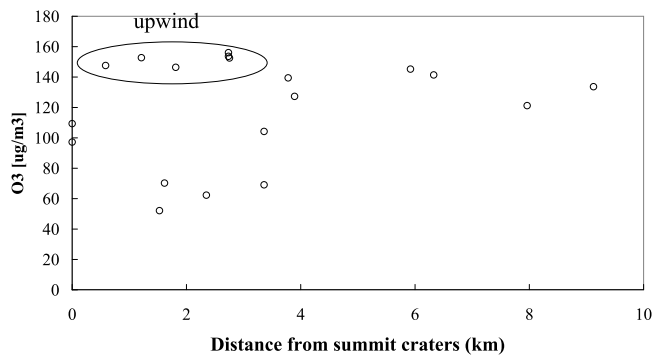
Figure 1. Measurements of ozone concentrations from diffusion tube sampling on Mt Etna in 2004. The upwind measurements are marked separately.

205 summit craters can be seen to remain in contact with the
206 slope for quite some distance before it disperses freely in the
207 free troposphere. We interpret the data such that O_3 deple-
208 tion can be observed in this region but not further away (and
209 below) the summit craters due to the plume no longer being
210 in contact with the ground. The two crater rim points have a
211 large uncertainty due to interference with SO_2 and should be
212 treated with caution. See Figure S1 in the auxiliary material
213 for a correlation plot of SO_2 and O_3, clearly showing that the
214 diffusion tubes were inside the volcanic plume.
215 [15] The O_3 measurements in 2009 are shown in Table 2.
216 The location is indicated as either on the slope or at the
217 crater rim of the South East (SE), the North East (NE) or the
218 Central (CC) crater. On 24 July, measurements were per-
219 formed at three different locations on the slope of the
220 Central Crater, for locations b and c two distinct time per-
221 iods with different O_3 concentrations could be identified.
222 At location c this coincided with a clear change in wind
223 direction, which might have affected the mixing of air
224 masses. It is unclear what caused the two distinct periods at
225 location b, but the 2nd time period at that location shows
226 significantly lower aerosol concentrations. It is interesting to
227 note that the period with lower aerosol concentration shows
228 a higher O_3 loss; this might hint at O_3 loss being more
229 efficient in more dilute plumes. The distance is given to the
230 crater that the respective plume originated from and the
231 transport time was calculated using locally measured wind
232 speeds (Kestrel 4500, Nielsen Kellerman) which showed a
233 very large variability which is reflected in the range of the
234 processing times given. O_3 was logged every 10s and the

variability of the O_3 loss was very large, therefore we show 235
the mean and the variability of the O_3 loss relative to 236
background concentrations. The available aerosol con- 237
centrations are listed in Table 2 as well. Please note that 238
above about N = 75 1/cm³ coincidence losses occur, so that 239
most in-plume samples are lower limits. This data clearly 240
shows that the measurements were taken inside the volcanic 241
plume. 242

3.3. Eyjafjallajökull

243

[16] Data collected during the 45 plume interceptions of 244
the Eyjafjallajökull plume are shown in Table S2 in the 245
auxiliary material. The plume transect time series for six 246
examples of such encounters are presented in Figure 2. Peak 247
O_3 loss ranged between 4 and 84% per transect, with a mean 248
of 37% and one standard deviation of 21%. There was 249
clear spatial overlap between the profiles of the O_3 loss 250
traces and those of the SO_2 in concert with a volcanogenic 251
mechanism for the observed O_3 depletion. This was aug- 252
mented with a marked correlation between the peak O_3 loss 253
per transect and the maximum values from the SO_2 sensor 254
(see Figure S2 in the auxiliary material) across the profile. 255
The presence of a volcanic plume in the region of depleted 256
O_3 was further confirmed by nephelometric measurements 257
of mineral ash, whose light scattering coefficient plume 258
profiles mimicked closely those of the SO_2 sensor. 259

[17] The scatter in the data likely arises from variations in 260
the plume composition at source and different ambient 261
conditions during the transport of the plume. Furthermore, 262
due to the prevailing anti-cyclonic meteorology in the North 263
Atlantic at the time of measurement, the plume was often 264
composed of mixed batches and filaments of different ages 265
due to recirculation, leading to variability in this processing 266
time. Variation in the chemical composition of the plume, as 267
a function of plume age, shows no discernable trend. 268

[18] The volcanic plume was found to be spatially inho- 269
mogeneous, being usually filamentary and composed of a 270
number of partially merged layers. Assessment of the age 271
of plume filaments intercepted is non trivial in the extreme. 272
For more details on our estimates of plume age, ranging 1– 273
9 days, see the auxiliary material. 274

4. Discussion and Conclusions

275

[19] The data from the three very different volcanoes 276
show that O_3 depletion is very widespread, starts within tens 277
of seconds transport time from the crater and is sustained for 278

t2.1 **Table 2.** Ground Based Measurements of O_3 Depletion in Vicinity of the Mt. Etna Summit Craters in July 2009[a]

Date	Location, Dist	Time	O₃ Loss	N [1/cm³]	A [cm²/m³]
16. July	background			17.5	6.3
09:53–10:17	SE slope, 1km CC	167–2000s	20 (2–38)%	562.2	22.7
11:51–12:35	NE crater, 2m NE	2–8s	24 (18–29)%	861.6	77.81
19. July				–	–
10:10–10:45	SE slope, 700m CC	108–125s	21 (−1–43)%	–	–
24. July	background			16.2	3.3
09:43–10:15	CC slope a, 770m CC	128–385s	22 (14–31)%	221.1	10.1
10:22–10:45	CC slope b, 590m CC	84–590s	33 (22–45)%	261.7	9.7
10:48–11:04	CC slope b, 590m CC	84–590s	40 (33–48)%	75.7	4.4
11:10–11:41	CC slope c, 526m CC	75–1052s	15 (8–21)%	146.2	10.0
11:43–11:56	CC slope c, 526m CC	88–526s	22 (16–27)%	–	–

t2.14 [a]"Dist" is the distance to the crater from which the plume originated (CC - Central, NE–North East), "Time" is the transport time
t2.15 from the crater to the measurement site, N is the aerosol number concentration and A the aerosol surface area in the size range
t2.16 probed. For more explanation see text. All times are local times (GMT+2h).
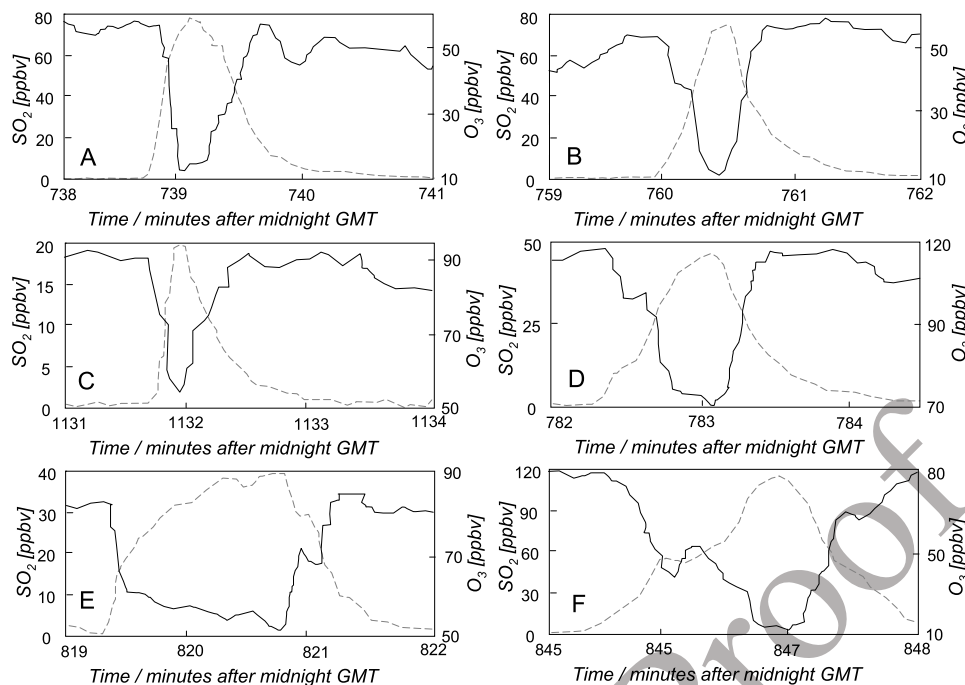
Figure 2. Airborne measurements of 6 interceptions of the plume of Eyjafjallajökull in 2010. Ozone mixing ratios are shown with solid lines and SO₂ with dashed lines.

279 several days. The model calculations by *von Glasow* [2010]
280 show a very rapid and sustained depletion of O₃ compared
281 to regions outside of the plume which is consistent with our
282 measurements. In the model the O₃ depletion is mainly due
283 to catalytic bromine reactions in the volcanic plume. The
284 self-reaction of BrO causes 84% of the O₃ destruction in
285 the first hour and 90% of the O₃ destruction in the first 6 h
286 after plume release. Clearly chlorine chemistry would be
287 another candidate for rapid O₃ destruction but the available
288 evidence hints at a larger role of bromine (see discussion by
289 *von Glasow et al.* [2009]).

290   [20] Ozone has been reported to be taken up on silicate
291 dust and has been suggested to be responsible for O₃
292 depletion in dust plumes [e.g., *de Reus et al.*, 2000]. Vol-
293 canic ash also contains large amounts of silicates so reactive
294 loss on ash might be an alternative or additional explanation
295 for the observed O₃ loss. The evaluation of laboratory data
296 by *Crowley et al.* [2010] recommends the following steady
297 state expression for the reaction of O₃ on mineral dust: $\gamma =$
298 $1500 \, [O_3 \, (cm^{-3})]^{-0.7}$, which for $[O_3] = 50$ppbv results in $\gamma \approx$
299 5.1×10^{-6} with a rather large uncertainty. It should also be
300 mentioned that γ is likely a function of time, depending on
301 the composition of the surface and ambient conditions such
302 as humidity.

303   [21] Assuming an upper limit for the heterogeneous loss
304 of O₃ of $\gamma = 10^{-5}$ the reactive aerosol surface area in the
305 volcanic plumes that we sampled would have had to be on
306 the order of 1.93×10^5, 3.2×10^3 and $130 \, cm^2/m^3$ in order
307 to explain an O₃ lifetime of 1min, 1h and 1day, respectively
308 (using equation (1) of *Crowley et al.* [2010]). Not very much
309 data is available about aerosol surface areas in volcanic
310 plumes but at Mt. Etna in 2009 we measured total aerosol
311 surface areas of only $4–78 \, cm^2/m^3$ (see Table 2). For
312 St. Augustine we estimated the aerosol surface area for the
313 day with the highest particle loading [see *Stith et al.*, 1978]

to be $\approx 150 \, cm^2/m^3$, which suggests that even in the most 314
dense parts of the plume that were sampled the lifetime of 315
O₃ to heterogeneous loss is on the order of a 1 day. Also 316
there is no clear difference in the O₃ loss as function of 317
estimated particulate mass flux (see Table 1), suggesting a 318
very weak influence of ash, if this influence is present at all. 319
Obviously the most dense parts of the plume had not been 320
probed so one might argue that in the early phases of strong 321
explosive eruptions the conditions for strong heterogeneous 322
O₃ loss might be given. As the dilution ratio in these plumes 323
is very large (one can make rough estimates from the change 324
in the SO₂ mixing ratio which near the vent is often several 325
ten percent but only on the order of a few hundred ppbv 326
hours to days downwind, see our data), ambient O₃ keeps 327
being mixed into the volcanic plume so in order to explain 328
sustained strong O₃ depletion hours and days downwind, O₃ 329
destruction must be ongoing. The surface areas required for 330
such a strong heterogeneous loss are not available, as 331
aerosol are also subject to very strong dilution and sedi- 332
mentation. Therefore we do not regard reactions on ash 333
aerosol as significant contributor to sustained O₃ depletion 334
in volcanic plumes. 335

  [22] It is very likely that ash is being processed in volcanic 336
plumes, for example by exposure to high sulphuric acid 337
concentrations. Literature data suggests that an upper limit 338
for the accommodation coefficient for the uptake of O₃ on 339
sulphate particles is $\alpha = 10^{-6}$ but likely much smaller 340
[*Sander et al.*, 2006] therefore our conclusion about the lack 341
of importance of heterogeneous loss of O₃ remains 342
unchanged. 343

  [23] Based on this we regard the most likely cause for the 344
observed rapid and sustained O₃ loss to be catalytic reac- 345
tions with halogen, mainly bromine, radicals. This is con- 346
sistent with observations of BrO both in the vicinity of 347
volcanic craters and several days downwind from the vol- 348

cano from satellites. The main features of the measurements are reproduced by the model by *von Glasow* [2010] indicating that our general understanding of O_3 chemistry in volcanic plumes as implemented in the model is realistic.

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