First we would like to thank the two reviewers who took a major part in the improvement of the article. We hope that our commentaries and modifications will fully answer to your questions and suggestions.

First reviewer :

Introduction, P8665, line 24. Volcanoes are not the only, and often not even the primary source of natural sulfate aerosol. For example, over ocean, DMS emissions can be the main source.

Thanks, we added the following sentence in the manuscript:

"The other main sulfuric acid precursors that generate fine sulfate particles are sulfur dioxide from anthropogenic sources and dimethyl sulfide (DMS), which is mainly emitted by marine phytoplankton."

2. Introduction, P8665, line 25. Although gravitational settling might be more rapid for volcanic ash than for sulfate aerosol, the relative impact of these species depends also on the relative optical depths, which vary from eruption to eruption, the particle absorption properties, and the likelihood of washout, which tends to affect sulfate more than dust.

Thanks. We added the sentence in the paper and slightly modified the introduction section.

"Although gravitational settling might be more rapid for volcanic ash than for sulfate aerosol, the relative impacts of these species depend also on their relative optical depths, the particle microphysical properties (i.e. size, absorption and shape) and the likelihood of washout, which tends to affect sulfate aerosols more than dust. The height of injection of these particles into the atmosphere is also an important parameter. When volcanic aerosols reach the stratosphere, their lifetime is significantly …etc"

3. Introduction, P8666, line 4. ": : :reach the free troposphere: : :"

Thanks, we included this correction.

4. Introduction, P8666, lines 19-20. In addition to the paper by Gasso (2008) that you reference, there are other papers, such as Scollo et al. (JGR 2012) and Kahn and Limbacher (ACP 2012), that use the visible part of the spectrum to characterize volcanic aerosol particle properties.

We added the following sentence:

"The data provided by the Multiangle Imaging Spectro-Radiometer (MISR) instrument were recently used to characterize volcanic aerosol particle properties in the visible part of the spectrum (Scollo et al., 2012, Kahn and Limbacher, 2012)."

We added Scollo et al., (2012) in the list of references.

5. Section 2.1.1, P8668, line 1. The retrievals use total and polarized radiance at 0.67 and 0.87

micron, plus 0.49 for the research algorithm. As the information content depends heavily on the input data, it would help to know also what range of scattering angles was included.

We added the following sentence:

"A large range of scattering angles (90-175°) was sampled by POLDER for the two case studies over cloud-free ocean scenes, which allows sensitivity to particles shape (Herman et al., 2005)."

6. Section 2.1.1, P8668, lines 6-7. Won't assuming that volcanic ash is non-absorbing in the visible cause the retrieved AOT to be underestimated? Ash absorption properties were reported from in situ measurements of these eruptions by Schumann et al. (ACP, 2011). I see you discuss the real refractive index from Schumann et al. in Section 4.

Right. We recall that we only use the operational algorithm to qualitatively detect the plume. The AOT and the absorption properties of the particles are depicted with the research algorithm.

It is mentioned in the paper: "…for a first qualitative detection of the volcanic plume …"

7. Section 2.1.2, P8669, lines 17-18. What effect on the uncertainties in the results does assuming the indices of refraction are spectrally invariant and the same for the fine and coarse modes, when the fine mode is probably dominated by non-absorbing sulfate and the coarse mode is mostly ash that has spectrally varying absorption?

We modified the research algorithm and reprocessed some data. We retrieved the complex refractive in the two modes. We retrieved the imaginary refractive index independently at 490 nm and 670 nm for the coarse mode (we assumed that the same imaginary values for the refractive index at 670 and 865 nm). The other parameters used to model the complex refractive index were independently retrieved and assumed spectrally invariant (i.e. real refractive index for the fine mode, real refractive for the coarse mode and imaginary refractive index for the fine mode). An example of the results obtained with this alternative algorithm is reported in table 1.

Alternative algorithm: 13 parameters to be retrieved	
AOT_865 nm	0.21 (0.02)
SSA_490nm	0.97 (0.02)
SSA_670nm	0.99 (0.01)
$r_{eff\ f}$ (microns)	0.26 (0.04)
$r_{eff\ c}$ (microns)	2.35 (0.36)
$m_{r\ f}$	1.47 (0.05)
$m_{r\ c}$	1.44 (0.05)
Fraction of spherical particles	0.23 (0.04)
Current algorithm: 10 parameters to be retrieved	
AOT865 nm	0.21 (0.01)
SSA_490nm	0.99 (0.01)
SSA_670nm	0.99 (0.01)
r_{eff_f} (microns)	0.24 (0.02)

r_{eff_c} (microns)	1.9 (0.2)
m_r	1.429 (0.005)
Fraction of spherical particles (within the coarse mode)	0.26 (0.08)

Table 1. AOT : Aerosol optical thickness, SSA : Aerosol Single Scattering Albedo, r_{eff} : effective radius, m_r: real refractive index. The suffixes _c and _f indicates coarse and fine mode parameters, respectively.

All the retrieved uncertainties increase, as expected. Note that the increase in the retrieved uncertainties is also (to a lesser extent) due to the additional errors considered for the surface (see responses to reviewer 2).

We analyzed more data with this algorithm. The spatial variability in the retrieved parameters increases. A surprising result is that the accuracy of the modeling is not improved and in fact decreases for most pixels, probably due to the existence of local minima. We think the problem is not well constrained (i.e. too much parameters to be retrieved for too few available information). We therefore conclude that the POLDER data (combined with the present method) can't be used to accurately retrieve the complex refractive index and its spectral variability separately in the two aerosol modes. Some assumptions must be considered.

Modifications of the research algorithm:

We however modified the research algorithm in order to improve our retrievals. The 490 nm spectral band primarily drives the retrieval of the aerosol absorption. We think that the retrieval of the spectral variability of the aerosol absorption requires the use of smoothness criteria to well constrain the spectral dependence of this parameter and avoid fluctuations. This technique is used in sophisticated aerosol retrieval methods such as the one described in Dubovik et al. (2011). This method is currently not implemented in our code. We therefore choose to assume a constant spectral behavior. We assume that the imaginary refractive index is 0.012/0.007 times smaller at 670 nm than at 490 nm. So, we still retrieve only one value for the imaginary refractive index but we now account for its spectral dependence. This ratio is based on the results described in Derimian et al. (2012). This study also found a rather small spectral dependence for the absorption between 670 and 865 nm and for the real refractive index of volcanic dusts. The same imaginary refractive index is therefore used at 670 and 865 nm and we still assume that the real refractive index is spectrally invariant. Spheroid models were previously used to compute the optical and scattering properties of the fine and coarse mode particles (i.e. we previously applied the same shape distribution for the entire particles size distribution). We now use the Mie theory for the fine mode particles. So, we now assume that the fine mode particles are spherical, which is expected for sulfate aerosols. This leads to minor changes and improvements in our retrievals. Figure 4 in the new version of the manuscript shows the "new" retrieved parameters and associated uncertainties for the near-source case. Most of our previous results remain. We still observe (as the distance to the plume decreases): (1) an increase in AOT, (2) an increase in the fine mode particles effective radius, (3) an increase in the fraction of non-spherical particles (within the coarse mode) (4) a decrease in the single scattering albedo, (5) a decrease in the Ångström exponent and (6) we still retrieve low values of real refractive index. We observe much less spatial variability in our retrievals than previously, especially for the retrieved aerosol single scattering albedo and fine mode effective radius. We have slightly larger values for the coarse mode effective radius than previously. We are still not able to model the POLDER data acquired over the thickest

part of the plume due to cloud contamination.

We added the following sentences in the paper:

"We made assumptions for the complex refractive index in order to reduce the number of retrieval parameters. This complex refractive index is assumed to be equal in both modes and the real part of the refractive index is assumed to be spectrally invariant. We assume that the imaginary refractive index is 0.012/0.007 times smaller at 670 nm than at 490 nm. This ratio is based on the results from Derimian et al. (2012), which analyzed the properties of a volcanic plume observed on 17 April 2010 using sun-photometer measurements. This study also found a rather small spectral dependence for the absorption between 670 nm and 865 nm. The imaginary part of the refractive index is therefore assumed constant between these two wavelengths in our algorithm."

"We only consider non-spherical particles for the coarse mode and we use the Mie theory to compute the optical and scattering properties of the fine mode particles. So, we assume here that the fine mode particles are spherical, which is expected for fine sulfate aerosols"

"The main retrieved parameters are … the fraction of spherical particles (FS), within the coarse mode,…"

8. Section 2.1.2, P8669, line 26, Table 1. On what are the assumed a priori AOT and particles properties for the fine and coarse modes based?

We added the following sentence in the paper:

"The minimum and maximal values used for the size parameters and for the real part of the refractive index are typical values observed for fine and coarse mode aerosols (Dubovik et al., 2002). The coarse mode and fine mode AOT used as a starting point for the retrieval are respectively equal to 0.4 and 0.05 (at 865 nm). These values were chosen based on the mean results obtained with the operational algorithm developed for POLDER. We use a value of 0.005 as a starting point for the retrieval of the imaginary part of the complex refractive index. This value is close to the one (0.004) prescribed by Schumann et al. (2011), which was derived from the joint analysis of ground-based lidar and airborne in-situ measurements acquired for volcanic plumes observed on April 16 and 17. For the imaginary refractive index we use an *a priori* of the maximal possible value of 0.02, which relies on findings reported in Derimian et al. (2012)."

9. Section 2.2, P8671, line 7. You might mention here that "AAC" means aerosol above cloud."

Ok, we included it.

10. Section 3.1.1, P8672, lines 16-17. Are there MISR stereo heights for any part of the 16 April plume? If so, they might complement the CALIPSO heights, because they cover more extensive areas.

There is no available MISR stereo heights data for the 16 Avril plume, at least not for the part of the plume that is described in our paper (the plume was largely spread over the Europe for this day).

11. Section 3.1.2, P8674, lines 11-16. At least for the upwind case on 7 May, the plume itself is as narrow as 10 km. Given the 6 km pixel size for POLDER, could some of the observed variability be due to retrievals done on pixels partly filled by the plume?

The plume was not so narrow for this day (at least, at the time of the POLDER overpass). The figure below shows the AOT retrieved by POLDER (without cloud mask). The green dots are the center of each POLDER pixel (native resolution ≈ 6 × 6 km^2).

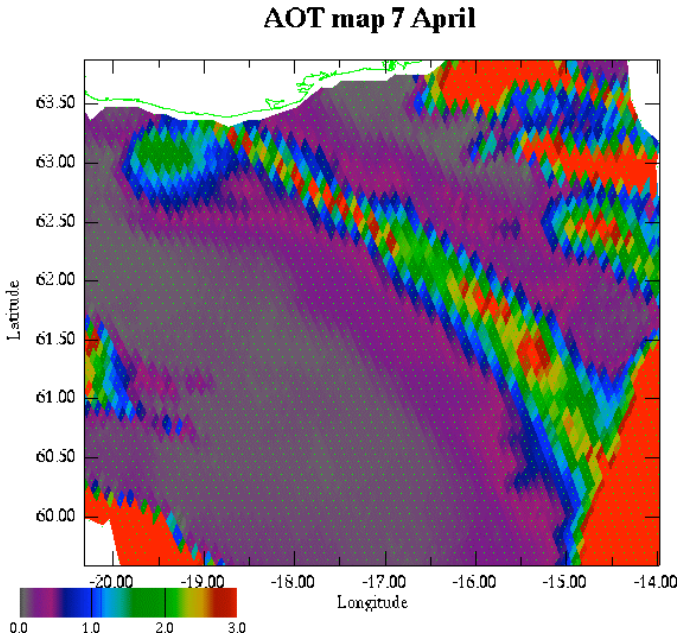

Figure 1

In the previous version of the paper, we used POLDER data aggregated at a coarse resolution (18 × 18 km^2) for the retrievals over ocean. Now, we show retrievals made at the finest POLDER resolution when we are looking at data acquired in the vicinity of the plume for the 7 April. We did not found much differences between the retrievals made at the coarse spatial resolution and at the fine spatial resolution. These additional retrievals now appear in figure 4 of the new version of the manuscript.

We added the following sentence in the paper:

"Note that the retrievals were performed at the finest spatial resolution of POLDER (6×6 km^2) in the vicinity of the volcanic plume …"

12. Section 3.1.2, P8674, lines 16-27 and Figure 3. What are the uncertainties associated with the retrieved particle size distribution, fraction spherical, and SSA values? This information would provide important perspective on which details of the retrieved particle properties, and especially which differences between retrieval cases, should be considered significant.

We reported in the text the mean retrieved uncertainties for both the coarse mode and fine mode effective radius and Angström exponent for the near source cases since this information are not shown.

We added the following sentences in the text:

"*The uncertainty associated with the coarse mode effective radius is quite large for the downwind case* and the differences observed between the two plumes for this parameter are therefore not significant."

"The real refractive index values retrieved for the downwind plume are too uncertain to drawn conclusions."

13. Section 3.1.2, P8675, lines 1-3, Figure 4. This is confusing. At what latitudes is the plume being sampled in Figure 4? Based on Figure 2, it appears that -20EÅNŽ longitude would be near-source, and -15EÅNŽ would be downwind along the plume. Yet the AOT increases, and the Angstrom Exponent, single-scattering albedo, and fraction spherical all decrease going from -20EÅNŽ to -15EÅNŽ, which seems opposite what might be expected, and the subsequent discussion seems to indicate that -20EÅNŽ is actually farther away from the plume core. So I guess you are sampling at some fixed latitude across the plume rather than along the plume. As such, this must be showing plume vs. background particle properties rather than plume particle evolution downwind. Please clarify.

Yes, we are sampling across the plume and yes we are showing plume vs. background particle properties rather than plume particle evolution downwind.

We already mentioned this information in the manuscript:

"Figure 4 shows the parameters retrieved for the near-source case as a function of the longitude. This allows us to observe the variability in the aerosol properties as a function of the distance to the plume."

We think that it is clear enough.

14. Section 3.1.2, P8676, lines 1-5. Could limitations in the assumed look-up-table particle properties, such as the particle shape model for volcanic ash, contribute to the surprising trends in the retrieved real refractive index and the fine-mode particle radius?

From Derimian et al., (2012) :

Dubovik et al. [2006] demonstrated very low sensitivity of angular dependence of aerosol phase function to the details of the shape of the aspect ratio distribution. Therefore, even if non-spherical volcanic ash may differ from desert dust in its distribution of particle shape, the utilization of the same spheroid mixture is not expected to produce important uncertainty.

Dubovik, O., et al. (2006), Application of spheroid models to account for aerosol particle nonsphericity in remote sensing of desert dust, J. Geophys. Res., 111, D11208, doi:10.1029/2005JD006619.

15. Section 3.2.1, P8677, lines 14-16. The quality of Fig. 5b could be better, especially in the region where the plume crosses the cloud bow; it is very small, and difficult to see features. Much of the foreground, where the glint dominates, seems unneeded.

Right, we modified the figure.

16. Section 3.2.1, P8677, line 23. It would be helpful to add the associated plot for typical cirrus to Figure 6a for comparison.

As suggested by reviewer 2, we added the reference of a study that describes the typical signature of polarized radiance from cirrus clouds (see responses to reviewer 2).

17. Section 3.2.2, P8680, lines 19-20. If you have a constraint on particle shape, in addition to particle size, this would strengthen the conclusion about whether the particles north east of the UK are volcanic ash.

The scattering angle sampled by POLDER was not favorable for this region (too narrow) and the fraction of non-spherical particles within the coarse was therefore not retrieved.

18. Section 4, P8682, lines 6-11. Since Kahn and Limbacher (2012) analyzed the 7 May and I see also part of the downwind plume on 16 April, is there any point in making further comparisons between the results of the two studies.

The downwind plume observed by POLDER on 16 April is not the same that the one studied by Kahn and Limbacher (2012) with MISR.

We already included some comments regarding the comparison of the results obtained for the 7 May plume by MISR and POLDER. We found a good qualitative agreement between both instruments.

19. Minor grammatical corrections scattered throughout: volcanic ash (not ashes), infrared radiation (not radiations), spectral behavior (not behaviors), particle absorption (not particles absorption), less than 5 x 10(-4) (not inferior to 5 x 10(-4)), etc.

Thanks, we included these corrections in the paper.