Response to Reviewer #2's comments on the paper entitled " Natural or anthropogenic? On the origin of atmospheric sulfate deposition in the Andes of southeastern Ecuador"

Please find here below our response to referee #2's comments. The text is also available as a supplement file (pdf) with different color keys for comments and answers to improve readability. Please note that "C" stands for comment and "A" for answer.

General comment:

C: The manuscript describes sources of sulfate deposition from five years of precipitation measurements at two mountain sites in south eastern Ecuador. The authors have brought together meteorological measurements, emissions estimates, and satellite data to compare to precipitation sulfate measurements in order to evaluate the impact of various S sources on remote mountain regions. This is a novel approach to a difficult problem; the identification of acidic inputs in sensitive remote alpine regions in Ecuador enhances the ability of researchers to apply similar techniques in mountain regions around the globe. Information on long-term changes in S inputs to remote alpine regions can be used to evaluate changes in anthropogenic versus natural emissions which in turn can be used to inform policy decisions. The manuscript was a pleasure to read. It is well constructed and aside from a few minor errors, typos and small grammatical mistakes that are described below, it is well written. The authors have been thorough in their description and the interpretation of the data is well argued and reasonably supported. Sufficient information is provided on the types of analysis and the potential pitfalls associated with the various data sets. The tables and figures are clear and appropriate.

A: We sincerely thank the reviewer for his comprehensive and positive general comments, the valuable recommendations and ther very accurate assessment of the text, figures and tables.

We have considered all reviewer's recommendations as listed below in detail. Because the manuscript has been edited, the lines indicated by the referee may not coincide with those were the changes were done. In the answers we are indicating where the requested changes were applied.

Specific suggestions:

C: Abstract: Line 11: been instead of being Line 14: conditions affects the origin...

A: Changes were made in lines 12 and 15.

C: Page 2: Line 8: effects were found to be more serious Line 10: only a few studies Line 31: contribute larger amounts

A: Changes were made in lines 10 and 12 and in page 3, line 5.

C: Page 3: Line 7: surveys in some (duplicate word) Line 11: local sources such as Line 12: deposition Line 19: were given special attention

A: Changes were made in lines 13, 17, 18 and 25.

C: Page 4: Line 1: emissions as accurately as possible Line 6: to determine sulfate deposition Line 17: It is unclear what is meant by "anthropogenic replacement systems" here. Please clarify. Line 23: are only a few sources

A: We applied the suggested changes in lines 8, 14, and page 5, line 3. In line 27, "anthropogenic replacement systems" were replaced by "pastures".

C: Page 5: Line 14: define MS (meteorological stations?). This acronym is described in a figure caption but it should be spelled out the first time it is used in the text. Line 18 and throughout the manuscript from this point forward SO2 – the 2 needs to be a subscript. Line 24: and transport to the observation sites.

A: changes were made in line 26, and page 6, lines 4 and 9.

C: Page 7: Line 4: that fires and volcanic emissions

A: changes were made in line 30.

C: Page 8: Line 23: pH

A: changes were made in page 9, line 21.

C: Page 9: Line 13: Before proceeding with Line 20: highest precipitation and OP inputs Line 23: observation period at around Line 25: spell out what the acronym MAD means here. Line 26: both types of precipitation input

A: changes were made in page 10, lines 9, 22, 25, 28 and 29.

C: Page 10: Line 1: highly loaded rain and OP – specify what the rain and OP is loaded with – ions?, sulfate? Line 6: generally higher when Line 26: from not form

A: changes were made in page 11, lines 6, 11, and 21.

C: Page 11: Line 8: and pass over the sources Line 12: using a cross-correlation Line 24: It is observed that Line 28: at this altitude Line 29: topographical locations

A: changes were made in page 12, lines 15, 19, and page 13, lines 3, 8.

C: Page 12: Line 17: Besides this there Line 23: airstreams Line 24: dark grey bars. Line 25: Contrary to this, during wind conditions...(light grey) Note that the light and dark grey description doesn't match the graphic.

A: You are right, I made a mistake in the text. The graphic is correct. I will switch "dark gray" and "light gray" in the text.
changes were made in page 13, lines 28, and page 14, lines 5, 6, and 7.

C: Page 13: Line 2: small rather than light Line 6: showed the same peak coincidences at el Tiro Line 8: contributes Line 11: small rather than light Line 12: are also higher than in rain here. Line 14: of the type of precipitation

A: changes were made in page 14, lines 14, 20, 22, 24, 25, and 27.

C: Page 14: Line 3: loadings in factors 1, 2, 4 and 5? Is the lack of factor 1 here a typo? If not, explain. Line 25: sources did not play Line 26: substantial

A: Thanks. Yes it is a typo. It should be "loadings in factors 1, 4 and 5". I actually realized that there was also an error in table 4. The last two row names in a) should be "Cerro del C. OP SO_4^-" and "Cerro del C. rain SO_4^-". In b) it should be " El Tiro OP SO_4" and "El Tiro rain SO_4^-".
changes were made in page 15, line 15, and page 16, line 11. Error in table 4 were corrected too.

C: Page 15: Line 1: north and

A: changes were made in page 16, line 18.

C: Page 16: Line 1: is relatively low here.

A: changes were made in page 17, line 20.

Response to Reviewer #3's comments on the paper entitled " Natural or anthropogenic? On the origin of atmospheric sulfate deposition in the Andes of southeastern Ecuador"

Please find here below our response to referee #3s comments. The text is also available as a supplement file (pdf) with different color keys for comments and answers to improve readability. Please note that "C" stands for comment and "A" for answer.

General comment:

This study conducts source-receptor relationships between collected wet deposition at two mountain sites and known SO2 emission sources. A large amount of work is presented and is worth to be published. The relative importance of precipitation and occult deposition need to be explained carefully due to the factors explained below.

A: We sincerely thank the reviewer for his positive general comments. The referee is right in bringing this point to the discussion. Occult precipitation at this altitude (2870-3200 m.a.s.l.) represents about 35% of the total precipitation in comparison with only 5% at 1800-2000 m.a.s.l. Furthermore, sulfate concentrations can be many times greater than in precipitation at the same site, what turns this type of deposition very important in mountainous areas with high cloud immersion frequency like the south-eastern Andes of Ecuador. Because of the higher wind speeds at this altitude cloud interception and wind driven horizontal precipitation is also more important as in lower altitudes, especially on summits and windward slopes .

Specific suggestions:

C: According to descriptions in Section 3.1.1, sulfate deposition collected in this study seems to include a portion of dry deposition (i.e., it is not a wet only collector). Apparently, dry deposition contributes a larger fraction of the total collected deposition in occult precipitation than in rain. It should also be noted that the actual dry deposition to forest canopies may be much higher than the portion collected by the instruments due to the larger surface areas of forest leaves. A brief discussion on this point and uncertainties caused in the experimental design should be added in this section and in places where total deposition amount is discussed.

A: It is true that we did not use wet-only collectors and therefore dry deposition is also adding to the total deposition. However, considering the weather conditions of the area, this sort of deposition should not play an important role. We added a sentence on this in page 6, lines22-28.
Certainly the deposition to forest canopies is not equal to that collected by the instruments and the relation between both catching efficiencies are an important parameter for evaluating aerosol inputs into the ecosystem. However, since our focus was on the characterization of deposition at two topographical sites and the identification of sources for atmospheric sulfate, the data from standard fog collectors was sufficient to fulfill our objectives (Ritter et al., 2008). We therefore did not calculate cloud/rain water interception by trees. Other sub-projects in our research unit are concerned with this and with the effects of nutrient and pollutant additions to the ecosystem (Homeier et al., 2012). We added a few sentences on the relation between catching efficiencies from trees and collectors in page 6, line 29 and page 7, lines 1-3.

C: Most receptor-based source-receptor relationship studies use measured concentrations at the receptor site. This study uses deposition data directly, and thus involves more parameters.

A: Our text was probably not clear enough, which has certainly misled the referee to what appears to be a misunderstanding. We did not use deposition data directly. For source-receptor relationship we calculated Volume Weighted Monthly Mean concentrations (VWMM, please refer to page 10, lines 12-14). Total deposition was calculated to have a measure of sulfate input variability per unit area, which is of importance for assessing the impacts on ecosystems and for nutrient manipulation experiments (e.g. NUMEX, Homeier et al., 2012 and Wullaert et al., 2010). We have rephrased some sentences in the whole text sometimes replacing "deposition" for "concentration" to make more clear what we actually did.

C: Sulfate wet deposition in rain includes two parts: in-cloud scavenging which likely related more to back trajectories and below-cloud scavenging which likely related more to local ambient SO2 and sulfate concentrations.

A: We agree with the referee in this statement. Below- cloud scavenging (rain deposition) is influenced by local concentrations as rain drops fall to the ground. In-cloud scavenging (direct deposition of pollutants incorporated by nucleation scavenging and collision into advected clouds onto the terrain; this is also called cloud interception) is less contaminated with local concentrations and as a result more representative of pollutant concentrations from more distant upwind sources. Please refer to Makowski Giannoni et al. (2013).

C: On the contrary, sulfate deposition in occult precipitation should mostly be related to local ambient concentration. These factors may help to explain the differences identified between rain and occult precipitation.

A: Occult precipitation is the supply of water to soil or to vegetation that is not by straightforward rain, and so, is not measured by conventional rain gauges. In our mountainous region it is principally generated by advected clouds (in high mountains even convective clouds) impinging the mountain tops and enveloping them (cloud interception / immersion), which is different from local fog formation, mostly occurring at lower altitudes, but with very low frequencies (Bendix et al., 2008). There is nearly no real radiation fog involved. That is why most ecology authors refer to the ecotype between 1800 and 3200 m.a.s.l. in the Andes as cloud forest. For this reason deposition from occult precipitation here is not a priori related to local ambient concentration, but most probably with pollutants advected by clouds / moisture from upwind sources.

References

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