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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Natural or anthropogenic? On the origin of atmospheric sulfate deposition in the Andes of southeastern Ecuador" *by* S. Makowski Giannoni et al.

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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, lines20-26. 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

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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 27.

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, line 6). 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).

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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.

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/14/C6602/2014/acpd-14-C6602-2014-supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 13869, 2014.