

Interactive
Comment

Interactive comment on “Amino acids in Arctic aerosols” by E. Scalabrin et al.

E. Scalabrin et al.

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We would like to thank the referee for the constructive and helpful comments. The time and effort spent by the reviewer on this paper are greatly appreciated. Our responses to each comment are given below.

P 1, I 4: “The wide range of amino acid reactivities suggest that amino acids may serve as markers of atmospheric transport and deposition of particles” Why? Could you explain this with one sentence?

Response: The combination of information derived from compounds with different reactivities is useful in understanding the aerosol age. Glycine has a half life >19 days in atmosphere and might be considered a tracer of organic nitrogen emissions; on the contrary methionine is highly reactive and it's rapidly destroyed in atmosphere (half life ≈2 hours) [1]. Moreover, the oxidation of this amino acid produce Methionine sulfoxide

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(MetSO). Met/MetSO ratio could be an indicator of aerosol age over short periods of time. The major product of MetSO oxidation is Methionine sulfone (MetSO₂) which could indicate a more aged aerosol.

P2, I 8: "Amino acids can be involved in cloud formation or act as ice-forming nuclei due to their hygroscopicity (Szyrmer and Zawadzki, 1997) and affect atmospheric radiation balance and climate (Chan et al., 2005)." This is true for all kinds of aerosols and does not justify why you have a special look at amino acids. Their share in the total aerosol mass is likely very small, so why should you study them? One reason could of course be that they are markers for certain types of aerosol sources but you did not explain which sources are represented by which type of amino acids.

Response: Water soluble organic carbon is widely recognized to have a role in cloud formation as important as inorganic species [2,3,4,5]. Despite this, this aerosol fraction is not well characterized. As demonstrated by Facchini et al. 1999 [6] the presence of organic solutes in aerosol particles can significantly lower surface tension, by up to one third; such changes, if they occur in droplets near the critical size for nucleation, lead to an increase in droplet number and then in cloud albedo. Moreover O'Dowd et al. 2004 [7] showed that the addition of a little mass of water soluble organic compounds in a inorganic aerosol population increases the cloud droplet number concentration by about 15-20%. Saxena and Hidelmann 1996 [4] suggested amino acid as a potentially important class of hygroscopic compounds, due to their low vapour pressure and high water solubility. Kristensson et al. 2010 [8] concluded that in most mixed particles of atmospheric relevance the amino acids will be effective CCN material. Chan et al. 2005 [9] measured the water uptake of seven amino acids, demonstrating that arginine and asparagine might act as efficient cloud condensation nuclei. Also Leucine has been shown to possess ice-nucleating ability [10]. Amino acids are recognized tracers of biological aerosol contributions, from marine and continental sources both, deriving from bacteria [11,12] and phytoplankton [13] associated with bubble bursting process [14,15], plants [16], debris and pollen [14]. Unfortunately specific sources are not

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well characterized yet and it's not well understood which amino acid represents which source. For this reason we need to deepen our knowledge about amino acids; Polar Regions offer a unique opportunity to better investigate such compounds as well as other water soluble organics.

P 2 I 16: "Free amino acids (FAAs) are particularly bioavailable and therefore may also significantly contribute to organic nitrogen and carbon in atmospheric depositions": How much is "significantly"?

Response: Amino acids accounted for the 20% of aerosol water soluble organic nitrogen in California [17], 19% of the total organic N in dry season aerosols and 2.5% in wet season aerosols from Brazil [16]; in rainwater collected at Wilmington, North Carolina, free amino acids represented 0.02-0.15% mole fraction in marine rain [18], 2% of the dissolved organic carbon [19], or 17% of organic nitrogen (2.55% of total nitrogen) [20,21].

P2 I 28: If they are detected in tobacco smoke, why does this show that they are associated with combustion-derived aerosols? The product of a combustion process depends on what is burned. I wouldn't agree that tobacco represents a number of different combustion processes.

Response: There are not clear evidences that combustion-derived aerosol can contain amino acids but some studies suggest a possible relation between this source and a few amino compounds. As we already cited in the text Mace et al. 2003 [16] suggested biomass burning as an amino acid source in the Amazonia Basin; Laskin et al. 2009 [22] proposed that Leucine, which is the most thermally stable amino acid, might survive to high temperature during biomass burning and detected a Leucine methylated derivative in aerosol produced by Pinus Ponderosa laboratory combustion. Violaki et al. 2010 [23] found correlation between Glycine and Elemental Carbon ($r^2=0.7$, $p<0.0001$) in PM1 collected in Finokalia, Greece, suggesting contribution from combustion processes to the glycine levels.

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P3 I 5: “During the Arctic summer, aerosol chemical compositions are strongly affected by local natural emission sources and aerosol concentrations are comparable to the Antarctic flux” So you expect that your summer measurements are mainly influenced by local sources. If this is true, why do you take 4-5 days back trajectories into account?

Response: Because we expect to find mainly local influences but we can't exclude other possibilities. We need long-term trajectory to verify if there was the contribution of long range transport from near continents and not only from the Ocean.

P3, I 8: “This increased input of FAAs from Eurasia has the potential to alter regional bioavailability of organic nitrogen as well as changing the formation of cloud condensation nuclei, and by extension, regional cloudiness and precipitation.” This sounds a bit exaggerated taking into account that amino acids are only a small fraction of the total aerosol. Could you say how big their share in the total aerosol mass is?

Response: Unfortunately we can't say how much amino acids contributed to the total aerosol mass because we can't measure Arctic aerosol mass and, to our knowledge, no data about the aerosol mass concentration levels at Ny Alesund and the Arctic region are available. In polar regions aerosol mass has rarely been measured due to the extreme climatic conditions and the low aerosol levels that require special laboratory rooms and apparatus to obtain reasonable precision and accuracy values; in addition, to ensure reliable results, several hours of measurement are needed, so more scientists should have been involved in the sampling campaign. Recently Annibaldi et al. 2011 [24] directly determine by gravimetric analysis the aerosol mass concentration in Antarctica, highlighting all the complexity of their work.

P3, I 12-23: Why did you choose these compounds? Why is it interesting to look at different size classes? Please explain these questions to the reader.

Response: We choose these compounds because they might be used as markers for bioderived aerosol and indicators of aerosol age, as we explained before. The analysis of different aerosol size classes is interesting to understand how amino acids are

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distributed and to point out particular tendencies: glycine for example has higher levels in the <0.49 filters; on the contrary Glutamic acid and Glutamine tend to accumulate in the 7.2-3.0 μm . Moreover, largest aerosol particles can't be transported over long distances, and then should derive from local sources. As we reported at P 8, I 11-22 larger aerosol fractions may be related to bubble bursting phenomenon while fine particles may be more affected by ageing and transport. We added in the text at P 3 I 19 :“Amino acids might be used as markers for bioderived aerosols and the different compound reactivity may be useful to determine aerosol age. The analysis of different aerosol size classes permits to highlight particular amino acid size distribution tendencies and may help the source apportionment study”.

P3, I 27/28: You should say here how long the sampling periods are.

Response: We added in the text at P 3 I 26:“Sampling periods varied between 131 and 246 hours”

P3, I 28: Why did you focus on summer? Just for practical reasons or because of a scientific hypothesis that is not mentioned here?

Response: We focused on summer because during this period the Arctic is not so much affected by Eurasian and American emissions, the number of sources is relatively low and the source apportionment study may be easier. In summer the contribution of natural sources is relatively higher so is the better period to study this kind of compounds, which are of mainly biogenic origin.

P4 I 23: Please explain why the filter is broken into pieces.

Response: Filters are broken in pieces to let the extraction solvent seep into the filters and obtain a high extraction efficiency. We added in the text at P 4 I 26:“to maximize the extraction efficiency”.

P4, section 2.3: What happens after the described steps? Which volumes were used for the final analysis?

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Response: We extracted the filters twice; slotted filters were extracted with 10 mL the first time and 5 mL the second one. The background filters were extracted with 35 mL the first time and 5 mL the second one. The two extracts of every filter were unified, filtrated through a 0.45 μm , Ø 25 mm PTFE filter and then directly analyzed in HPLC-ESI MS/MS. For the analysis we used an injection volume of 5 μL .

P5, section 2.4: What are the recovery rates of the standards?

Response: The recovery rates of the standards are reported in the references cited in the text, where the method is extensively described [25,26]. The recovery rates of the internal standards can't be calculated.

P5, section 2.5: What were the concentrations in the blanks? You refer only to the standard deviations. What is the relation of the LOD to the minimum detectable concentration in air?

Response: I added in the Table 1 the LOD values in terms of fmolm-3 and the average blank concentrations in ng and fmolm-3 both. We add in the text at page 17373, line 3: "The LOD values in terms of fmolm-3 were calculated using the averaged sampling volumes, 11982 m3 and 11613 m3 for slotted and background filters respectively".

P 5, I 30: "Of the 17 collected samples, 9 were analyzed in all 6 of the size ranges. For the other 8 samples, only the background (<0.49 μm) filters were analyzed." Please give the reason for this.

Response: We choose 9 samples (more than a half of all those collected) to be analyzed in all stages, in order to obtain a representative number of sample for all the summer period. We expected to find the highest amino acid concentrations in the background filters (so we consider all those samples) due to the elevated particle number concentration in this size range during the summer [27] and the significant amount of ion mass and biogenic compounds detected in submicron Arctic particles [28,29].

P6, I 3: "Samples collected during the first week of May have the highest concentra-

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tions, with total FAA concentrations double than those collected from June 6-16.” Why do you compare these two periods?

Response: As shown in Figure 3, from June 6-16 we detected the highest amino acid concentrations in all size aerosol fractions, excluding sample collected between May 13-19 that we considered influenced by the volcanic ash transport. We compared the sample collected during 1-7 May, which has the highest amino acids content, and the sample collected during 6-16 May, which is the most abundant sample collected in “ordinary” conditions.

P6, I 11-19: Which conclusions can you draw from a comparison to other observations reported in the literature? Are the amino acids of marine origin?

Response: Our amino acid relative abundances are similar to those obtained in aerosol sampled in remote [13,30] and marine areas [23]; however, in these studies, the authors suggests that continental contribution can’t be totally excluded. What we can say is that, in general, our observations better agree with what found in marine areas than in continental ones.

P 6, I 19: What is meant here with “profiles”?

Response: For profile we mean that all the amino acid relative abundances are similar in all samples.

P 6, I 33: So virus and bacteria are rather big. You need to explain this here.

Response: We add in the text, p 17374, I 20:”They attributed the fine mode ($<1 \mu\text{m}$) to the presence of organic surfactants or to the influence of gas-to-particle conversions and the coarse mode ($>1 \mu\text{m}$) to the presence of live species, especially bacteria that typically range in length from 3.7 to 7.5 μm in tropical areas or 0.5 to 3 μm in the Arctic [32]”.

P7, I7: If the large particles originate from bubble bursting, why don’t you see them all the time? Is there any hint that bubble bursting was particularly dominant as a source

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of amino compounds connected with large particles during these two periods?

Response: We don't see largest particle (10-7.2 μm) all the time because these are probably freshly emitted, deriving only from very close sources; as we already explain in the text (p. 17375, I 7) this kind of particles can't be transported so far and tend to settle gravitationally [33]. As you can observe in Figure S1a and S2d in the Supporting Material, corrected as you recommended us, the transport pattern of these two samples don't differ from the others. As suggested by previous studies [34,35] we can hypothesize that particles in this largest fraction (10-7.2 μm) can mainly be attributed to bubble bursting process. In the last discussion paper version, the one published in ACPD, you can find a more extensive explanation about this point than in the version you refer to.

P 7, I 12-20: What does this tell me? Can you draw any conclusions from this?

Response: We can't compare our findings with other studies so we can only describe what we found. Size-segregated analysis, as reported in the latest paper version published in ACPD, at p. 17376, I 3, allow us to point out some amino acid distribution tendency in specific fractions. Similar suggestions, to our knowledge, were never reported before.

P 8, I 8-11: Again my question is if tobacco smoke is representative for biomass burning in general.

Response: See the reply to comment P2, I 28.

P 8, I 14: you should not call volcanic ash "smoke".

Response: We replaced smoke with "ash"

P8 I 12-26: I would doubt that this is a proof that FAAs from Eyjafjallajökull were detected. Do you have any other indication that volcanic ash was in your samples? If you calculate the trajectories for arrival heights of 2500, 5000 and 7000m: What does this tell you? These air masses were not detected at 500 m altitude where your

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sampler is located. Were ultrafine particles emitted from the volcano or what are the corresponding precursor gases that stem from the volcano? The back trajectories may indeed help you to interpret your results but I have the impression that you used them in a wrong way. Why do you have only one trajectory for the entire sampling period? Is this the trajectory that arrives at the sampling site at the end of the period? If you collect for 131 to 246 hours you need to take all trajectories that arrive during this time at your sampling point into account. It might be sufficient to consider 2 trajectories per day, but you cannot use just one for the entire period. This is a severe problem for your entire interpretation. You need to solve this.

Response: We corrected the backward trajectories as you suggested us and we thank you of your helpful observations. We calculate two trajectories per day for the entire sampling period at altitude of 500 m. As shown in Figure 4c, the back trajectories of the 13-19 May sample demonstrate a clear contribution of air masses transported from the Icelandic area, where the Eyjafjallajökull volcano was erupting since the 3rd of May. Also in the sample collected between the 1-7 May a back trajectory could indicate air masses transport from the Iceland to the Svalbard during the 7th of May. In this case, however, the altitude of the trajectory can't encompass the atmospheric height of the volcanic plume [36]. We then suppose that the volcanic eruption might have affected the amino acid levels in the 13-19 May sample but not in the others. Any studies on the influence of the 2010 Eyjafjallajökull eruption in the Arctic region haven't been published yet, but some results have already been presented at meetings and conferences. At the 10th Ny Alesund Seminar Kriews et al. 2011 [37] shown a study on the chemical characterization of the volcanic plume at Spitsbergen (Svalbard Islands) at the ground level, in aerosol and depositions both, verifying strong differences in element concentrations and compositions before and after the volcano ash plume reached the sampling location. Moreover, remote sensing images from IAVCEI (International Association of Vulcanology and Chemistry of Earth's interior) [38] demonstrate that from 17th of May to 19th of May the aerosol levels in the Svalbard increased dramatically, caused by ash and gas transport from the Iceland. For these reasons we consider our

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conclusions reasonable. With respect to the particle size emitted from the volcano, as reported by Schumann et al. (2011) [39], the ash plume contained ultrafine particles ($<0.5 \mu\text{m}$) in the major part ammonium nitrates and sulfates of secondary origin, on May the 2nd and 17th both. A minor part were composed by mixed, silicatic and quartz particles. We change the text at page 17377 L 17-21 and 17377 l31-p 17378 l 5.

P9, l 5: "...suggesting that during the Arctic summer regional aerosol concentrations are significantly less than boreal winter and spring concentrations." What is the reason for these differences? Is it due to transport patterns or due to missing sources?

Response: During the winter season the Arctic lower troposphere is well isolated because the isentropes form a closed dome over the Arctic. This situation, associated with the total darkness, causes the inefficiency of removal processes (precipitations, wind, dispersion, photochemical reactions) of aerosol and precursor gases. Although the polar dome creates a transport barrier (the polar front), this barrier is permeable in winter[40] . The mean circulation in winter is characterized by low-level transport from northern Eurasia across the Arctic toward North America. On the contrary, in summer, the solar radiation causes the thermal inversion of the Arctic troposphere, determining the dispersion of pollution. In summer, the transport pathway is directed from the North Atlantic Ocean across the high Arctic toward the North Pacific Ocean, and the transport is only half as fast as in winter. Because of the slower transport and more-efficient removal processes in summer, pollution concentrations are lower and high-latitude sources of air pollution become more important than in winter [41].

P9, l 12-14: Generally, one would expect that the longer the aerosol particles stay in the atmosphere the more they will be found in larger size classes and in the accumulation mode with a size of about 1 micrometer. How does that fit with your observations?

Response: We found the higher amino acid concentrations in the Aitken and Accumulation modes but we can't directly relate this with aerosol concentrations. Probably these two aerosol size ranges represent an important part of total aerosol particles.

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With respect to the larger particles, only in some samples, as described at p 17374 I 24- p 17375 I9, we found relevant amino acid concentrations in this size fraction.

P9, I 16: What is the half life of MetSO₂ ?

Response: To our knowledge, no studies about the atmospheric half-life of MetSO₂ have been conducted. Karunakaran-Datt and Kennepohl (2009) [42] concluded that MetSO₂ is oxidatively and reductively inert in photolysis conditions and that its formation seems to be irreversible.

P9, I 20: “The mean Met/MetSO ratio was 0.4 ± 0.1 indicating the contribution of mid-distance (10s to 100s of kms) aerosol sources.” How exactly do you come to this conclusion?

Response: Considering the Met half life, the 0.4 ratio between Met and MetSO is obtained in more or less 5 hours. Looking at the back trajectories, which give indications about the transport speed, we can see that in about six hours (all the repeated marks in the trajectory lines) the air masses can move of about 10-100 kms.

P10, I 19: “Tyr and Ser are reactive and hydrophilic compounds, while Pro and Val are hydrophobic, so the relation between the four compounds appears to be more affected by source differences than by similar chemical behaviors.” I do not understand this point. Why do they cluster if their sources and their chemical behaviour is so different?

Response: We don't know exactly why they cluster, we can just report what we observed and hypothesize that they may derive from different sources and mix in the atmosphere during the transport. At the moment we can't identify specific sources for these compounds.

P11: Conclusions: It would be good to know which of the “classical” compounds were found in the aerosol samples, e.g. sea salt, nitrate, sulphate, etc, to facilitate the interpretation of the results. I do not agree that you proved and influence of volcanic activity for May 13-19 period.

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P12, I 2: “Numerical simulations demonstrate that the sea spray height and transfer of local sea spray aerosols to Svalbard depends on surface wind speeds (Doernbrack et al., 2010) suggesting that marine FAAs may increase during times of increase wind speeds.” Did you see this, too? I assume that wind speed information is available.

P12, I 7: “...although many of these originally marine aerosols contain aged sea salts that have reacted with atmospheric NO_x or HNO₃.” This was not shown in the paper before, although it would be of interest to know more about it.

Response: Unfortunately we don't have available data about inorganic aerosol constituents and wind speed. Surely it will be interesting to study and compare these results with our previous findings reported in this study.

Table 1. Table2: Why is MetSo not given in Table 1?

Response: We added MetSO in the two tables.

Table 2: Why is the mean for Ser given as 3×10^2 while the median is 262? How can the mean be less accurate in terms of valid digits than the median?

Response: As we know from statistics, the median is described as the numerical value separating the higher half of a sample or a population from the lower half. So 262, as all other medians, is the middle number of the group when they are ranked in order and for this reason is more accurate than the mean.

Figure 3 and 4: As mentioned earlier: I do not think that this analysis has been done in a proper way.

Response: We changed these Figures as you indicate us (see Figure 4 and FigureS1, S2, S3, S4).

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Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/12/C8397/2012/acpd-12-C8397-2012-supplement.pdf>

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 17367, 2012.

ACPD

12, C8397–C8417, 2012

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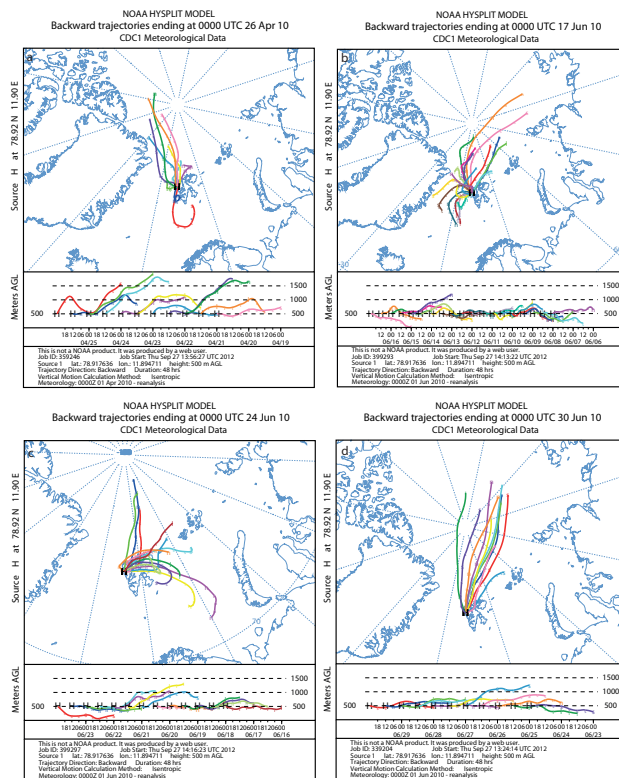


Fig. 1. Figure S1a,b,c,d: Back trajectories of the 19-25 April (a), 6-16 June (b), 16-23 June (c) and 23-29 June (d) samples. The 48 hours back trajectories are calculated at an altitude of 500 m.

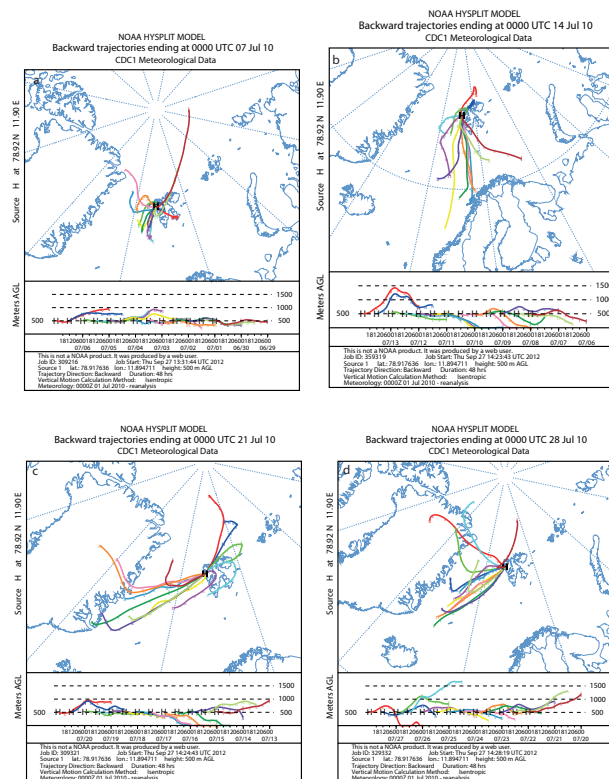


Fig. 2. Figure S2a,b,c,d: Back trajectories of the 29 June-6 July (a), 6-13 July (b), 13-20 July (c) and 20-27 July (d) samples. The 48 hours back trajectories are calculated at an altitude of 500 m

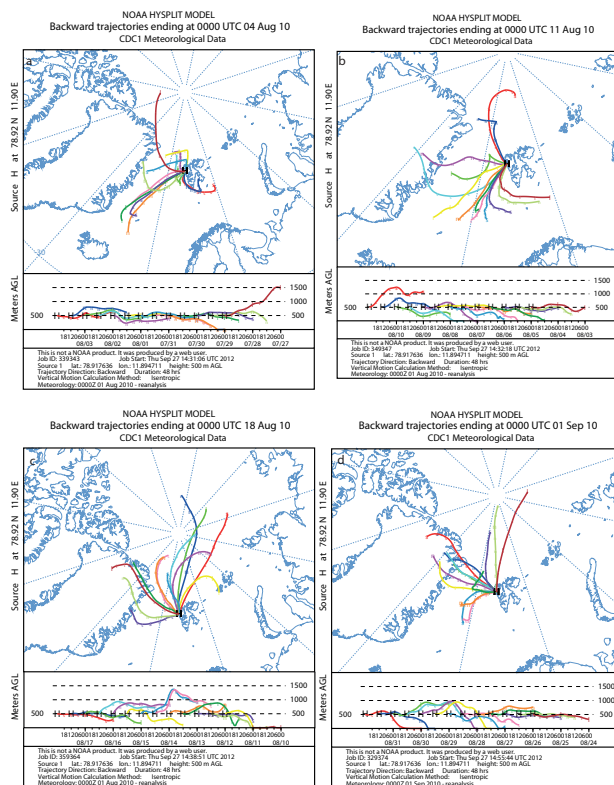


Fig. 3. Figure S3a,b,c,d: Back trajectories of the 27 July-3 August (a), 3-10 August (b), 10-17 August (c) and 24-31 August (d) samples. The 48 hours back trajectories are calculated at an altitude of 500 m

NOAA HYSPLIT MODEL

Backward trajectories ending at 0000 UTC 15 Sep 10

CDC1 Meteorological Data

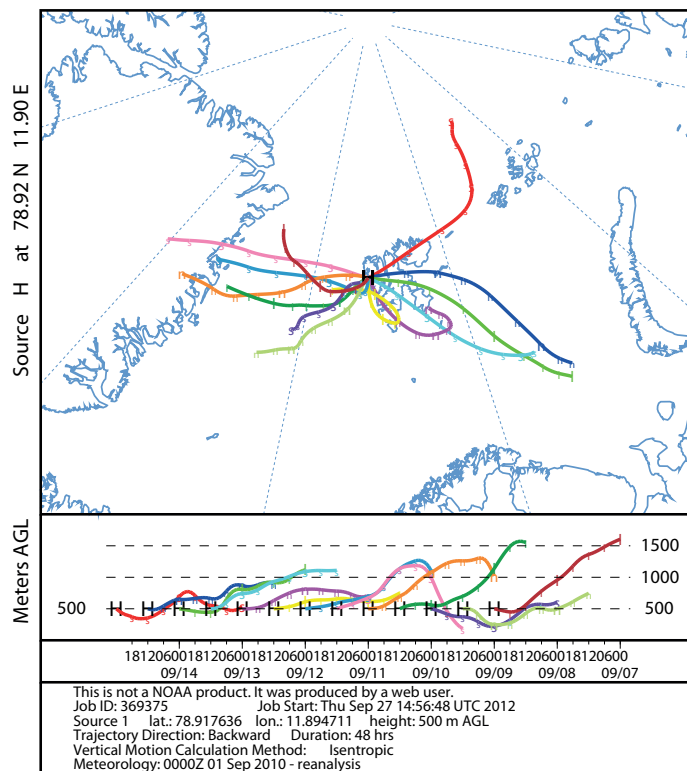


Fig. 4. FigureS4: Back trajectories of the 7-14 September sample. The 48 hours back trajectories are calculated at an altitude of 500 m for the entire sampling period, starting a new trajectory every 12 hours

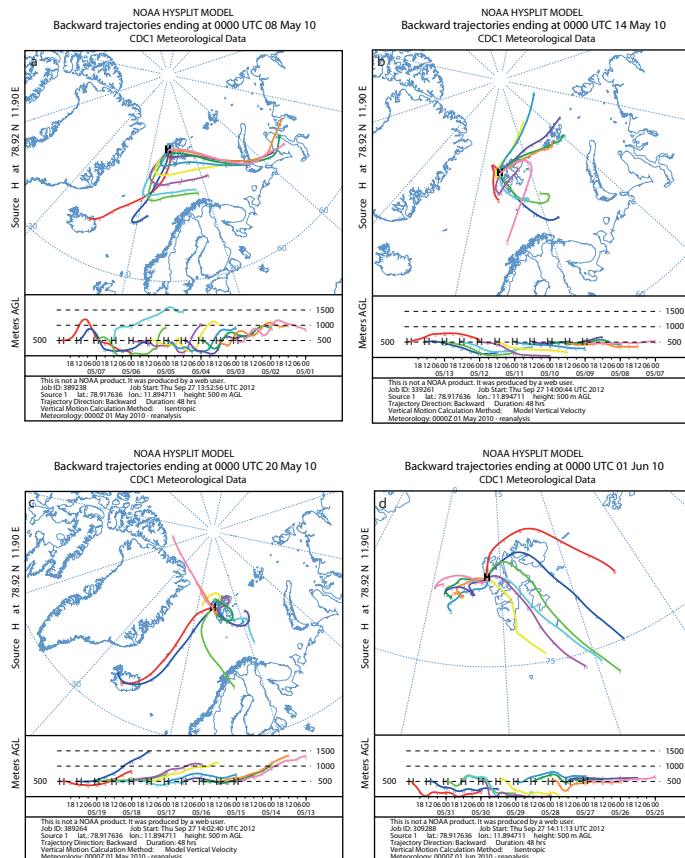
Interactive
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Fig. 5. Figure 4: 48 hours back trajectories of the 1-7 May (a), 7-13 May (b), 13-19 May (c) and 25-31 May (d) samples collected during the eruption of the volcano Eyjafjallajökull.

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