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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Transport of anthropogenic emissions during ARCTAS-A: a climatology and regional case studies" *by* D. L. Harrigan et al.

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Author Responses

"Transport of anthropogenic emission during ARCTAS-A: Mean transport characteristics and regional case studies"

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REFEREE #1

1. In section 2.2 (P.5441 L.4-5) It is explained that trajectories represent the paths taken by air parcels over a period of time. However, in numerous places in the text

this relationship between air parcels and trajectories is very confusing. In its current form the paper almost treat trajectories as a synonym for air parcels which is misleading. Please take this comment under consideration and rewrite these parts of the text. P.5436 L.13, P.5441 L.20, P.5441 L.26, P.5444 L.1, P.5448 L.2, P. 5451 L.5, P.5453 L.23, P.5454 L.16,

We have deleted the word “air parcels” and substituted “trajectories” at all locations in the text.

2. P.5438 L.19: Consider mentioning of the cooling effect of snow as well as of ice.

We have added this comment.

3. P.5439 L.21: biomass emissions -> should it state: biomass burning emissions?

We agree with your more precise wording and have made the change at every location in the text that is appropriate.

4. P.5441 L.8-10: Why were trajectories more appropriate than calculation by a Lagrangian particle dispersion model (LPDM)? Transport statistics have been calculated using LPDM’s in several studies in the past.

We are familiar with the LPDM (e.g., FLEXPART) and the references that the reviewer cites about it. In fact, we used it when writing the ARCTAS meteorological overview (Fuelberg et al. 2010, ACP). As a side note, we have now switched to the WRF-Chem CTM in our ongoing research.

We did use FLEXPART during the research presented here. It was used as a preliminary tool to help select the three individual case studies. Specifically, we looked for flights when plumes of anthropogenic CO from FLEXPART intersected the flight tracks. We then determined the particular leg of the flight when this occurred and began to analyze the chemistry and use trajectories to further explore the meteorology. There are several reasons why we did not use a LPDM in the current manuscript.

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1) Fuelberg et al. (2010) used trajectories to describe the overall northern hemispheric Arctic transport during ARCTAS. In the current manuscript, we break down that overall transport into components arriving from Asia, North America, and Europe. We thought we should use the same procedure here as before to allow readers to make an easy comparison between the overall and region specific results.

The LPDM is especially useful when considering what the concentration of a species is at a certain downwind location. That is possible because a specified release rate can be simulated by selecting the rate at which particles are released and then assigning an appropriate mass to each particle. However, the current manuscript only deals with transport pathsâĂŤnot the concentration at a given point. Therefore, the LPDM approach did not seem necessary.

2) The current manuscript describes three individual case studies. We wanted to know the origins and paths of air sampled along specific legs of certain flights. While FLEXPART can be used in a backward mode, I have always found it difficult to interpret and explain the backward FLEXPART results; explaining backward trajectory results is much simpler.

As noted above, we have used both FLEXPART and trajectories during this research. The important point is that our comparison of results indicates that the use of FLEXPART would not change any of the conclusions in the current manuscript. We have modified this section of text to better reflect the ideas stated above.

5. P.5455 L.26-27: Consider to reformulate this sentence. It is at the moment unclear if it should state that 54% of all trajectories calculated from releases between 40 and 50 degrees North reach the Arctic, or if it instead should express that 54% of the trajectories reaching the Arctic originate from releases between 40 and 50 degrees North.

Your second option is correct. We have rephrased the sentence to make that clear.

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REFEREE #2

1. P.5441 L.8-10: “Although . . . , trajectories were more appropriate for the applications. . . ” Please briefly explain in text why trajectory analysis is more appropriate for this study than Lagrangian particle dispersion models.

See our response to point 4 of Referee 1 (above).

2. P.5441 L.21-23: As I understand, trajectories were released daily at 2 pm from areas of interest. Why specifically 2 pm was chosen? Would it be a better way to derive a transport climatology using trajectories released, for example, 4 times a day?

We chose 2 PM LDT because it is near the time of maximum temperatures and when the planetary boundary layer often is deepest and turbulent mixing (parameterized in WRF) is generally most pronounced. Thus, it is when pollutants are most likely to be carried to higher altitudes where the winds are stronger and the trajectories are more likely to reach the Arctic within 15 days.

We have added the above reasoning to the text and state that the choice of 2 PM probably results in longer range transport than at other times.

3. P.5443 L.19-20: Please explain why ozone depletion episodes at low altitudes were excluded from this study and how it would affect the results and conclusions. For the discussion in question we were trying to determine concurrent local background values for each trace gas of interest. We eliminated stratospheric air because this was not representative of the local background. Similarly, part of the ARCTAS-A mission sought to understand halogen chemistry, and therefore ozone depletion events (ODEs) at low altitude were actively sought out. However they were excluded from the selection of background air because they were not considered to represent the composition of typical background air, plus these events were relatively oversampled and their inclusion could potentially skew the background values. Our reasoning was that this paper seeks to find urban/industrial and biomass burning signals enhanced over the local

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background, rather than enhanced over a combination of the local background plus air influenced by halogen chemistry. We believe that the exclusion of ODEs provides us with the most appropriate suite of background values to use at these latitudes.

4. P.5444 L.20-21: ". . . , and one distinct pathway is evident." Based on my visual inspection on Fig. 4b, I could not clearly identify the mentioned pathway. And it's difficult for me to identify pathways from Fig. 9b for North America and Fig. 13b for Europe too. Would it be possible to improve these figures to make pathways more distinguishable?Although it's challenging to show pathways in one single plot, the authors may want to try reducing the dot size or providing individual plot for each pathway as supplementary material to this paper.

Depicting the information in Fig. 4b and its counterparts was a real challenge. We tried several alternatives, and none seem any better (most were worse) than the current version. We believe the best approach is to tone down some of our statements (e.g., the pathways are not that distinct). We also now state how to interpret the resultsâĂŤthe orientation of the red and blue areas is a dominant pathway. We believe these modifications achieve your goal.

5. P.5447 L.1-6: Why C2Cl4 and HCFC-22 were excluded from the correlation analysis?

The four compounds included in the correlation analysis (H-1211, OCS, 1,2-DCE and CH3Cl are the four that Barletta et al. (2009) originally cited as tracers for air originating from China. Therefore we used these compounds to test for Chinese source influences. To further test for the urban/industrial nature of this air, we then folded C2Cl4 and HCFC-22 into the discussion. To address the reviewer's comment we have added the following sentence to the text at P5447 L6: "In particular, C2Cl4 showed very good correlation with the four tracers of air of Chinese origin (H-1211, OCS, 1,2-DCE, CH3Cl), with r2 values between ranging from 0.79 to 0.99."

6. P.5453 L.28-P.5454. L.1: "Although a few . . . , the majority . . . " Please quantify

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the numbers of trajectories in the above two categories.

We have added these numbers

REFEREE #3

1. It is not clear why the authors go through the exercise of double nesting the WRF simulations, this is never discussed. It is also not clear why those particular nests are chosen, they do not coincide with the trajectory starting locations.

As stated on page 5440, lines 18-20, the inner grids are “centered on three historically [wording now changed] significant anthropogenic source regions including Europe, Eastern Asia, and North America”. The nests do not agree exactly with the trajectory release locations (Fig. 2), but they are close. We used nesting to resolve these emission regions at the highest resolution that our computing capabilities allowed. We did not have the resources to use 5 km resolution over the entire domain.

Here is what the AMS Glossary of Meteorology states about nested grids: “A high-resolution region of discretization embedded within a low-resolution region of a numerical model or analysis system.

Fine resolution may be desired to focus on a mesoscale feature such as a tropical cyclone or on a geographical area of interest. In two-way nested grids, information is passed back and forth between the high- and low-resolution regions, in contrast with one-way grids in which information is passed merely from low- to high-resolution regions."

We would have used a 1 km resolution over the entire domain, but could not because of computing limitations. The guidelines for using WRF suggest that successively finer resolution nests should be at 1/3 the spacing of the larger (outer) grid. That is what we did (45, 15, 5 km).

We have modified the discussion on pages 5440 and 5444 to make these points clearer.

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2. A major limitation of the study is the use of 1x daily trajectory calculations on a 2x2 degree grid. This weakens the statistics, and could bias results by examining one particular time of day only for each area. The analysis of general transport characteristics should be redone with at least 4x daily trajectory calculations. Considering that the WRF model output is hourly, this should not be difficult.

See response 2 to referee 2 above.

3. It is unclear what is shown in Fig. 2, panels b,d,f. The CGRER data set is supposed to show anthropogenic CO emissions. But over Borneo for instance Fig. 2b shows as high emissions as for Japan. On panel 2f there are large CO emissions over equatorial Africa, higher than over central Europe, which is rather counterintuitive since these are not heavily industrialized areas. More details on the data set should be given, and a comparison with some other emission inventory be made.

The global CO emission distribution from the CGRER ARCTAS emissions show the same spatial distribution when compared to other global inventories including IIASA global emissions used in UNEP ABC report (UNEP BC report reference) and EDGAR. The CO emissions in Borneo are similar to those over non-urban parts of Japan and in Equatorial Africa compared to Europe are "correct" and show up in all current CO inventories. This reflects the fact that emission in areas like Borneo and Equatorial Africa reflect extensive use of inefficient combustion for residential cooking, while combustion efficiencies in places like Japan are very large –thus low CO emissions.

4. There is a significant amount of redundancy in Figs 6, 11, 15: Panels a,b can be condensed into one, and the boundary layer segments indicated by symbols or a more distinct color. This would provide space for bigger, more readable figures.

We agree that there is some redundancy in panels a) and b) of these figures, but do not believe that your proposal would achieve the desired result. There are many trajectories in panel a) and we fear that combining it with panel b) would lead to a real hodgepodge of information that would be difficult to see.

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Interactive Comment

We believe that both panels should be kept, and all panels should be made considerably larger. The versions you saw were about 40 mm wide, but a column in ACP is about 80 mm wide. Thus, the panels can be enlarged considerably and we trust this will be acceptable.

5. The sea level pressure figures in Fig. 7,8,12,17 are not very useful because they cover a too large area. It would be much more useful to show just one panel for each case when the air parcels supposedly are lifted up in a WCB, zoomed to a reasonable region. Other variables than SLP, for example cloud cover or precipitation, could be shown to strengthen the point of a WCB being present in the WRF simulation.

We have reduced the areal coverage of these figures so they now are larger and more legible. However, adding another parameter to the figures would make them very "busy". We believe that our use of the Eckhardt et al. criteria, together with the locations of the trajectories with respect to the surface cyclones provide convincing proof that WCBs are involved.

6. The writing should carefully distinguish between air parcels, air masses and trajectories. In the manuscript these terms are wrongly used interchangeably.

We have done this. See comment 1 of referee 1.

7. The method description of the trajectory calculations needs more details and clarification.

Page 5441 lines 4-7 gives a few basics of the trajectories and provides three references for more information. However, to address the reviewer's comment we have added more details on that page.

8. It should be more clearly stated in the manuscript that the paper is focused on case studies. I don't think that the term "climatology", even though in quotation marks, can be used justifiably for a one-month period. In particular the title of the manuscript is misleading in that sense, and the word climatology should not appear here. You

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could describe the month-long analysis as "mean transport characteristics during the ARCTAS campaign", or something along those lines.

We have changed the title as requested and modified the text to eliminate the use of “climatology” and have substitute “mean transport characteristics”.

9. It should be discussed that the present setup does not take into account that air masses can leave the Arctic again right after having crossed the 70_N boundary. Whether air masses actually remain in the Arctic for an extended period of time could be assessed with forward trajectories from the aircraft locations.

We mentioned this point on page 5444 line 17, however, we now amplify on the implications in the following sentences. We did not think residence time was pertinent to our study and did not investigate it

10. The authors suggest that in all three cases of interception of polluted air masses WCBs played a decisive role in the vertical transport behavior. While this may be a coincidence, this aspect should be discussed in more coherence in a separate discussion section. Is there actually at all a mechanism that could bring mid-latitude polluted air to the altitude of the aircraft within a handful of days, other than WCB uplift?

While WCBs were important in the three cases that we presented, we agree that it probably was a coincidence. We have added sentences describing mechanisms besides WCBs to the Summary.

11. The WCB criterion is explained 3x in each case study. Instead, it should be described 1x in the methods section. A table would be helpful to summarize the WCB criterion results.

We have consolidated all of the Eckhardt et al. WCB criteria into a new methodology section (new Section 2.3). Therefore the criteria are now only stated once. However, we still must refer to those criteria somewhat when discussing the individual cases.

12. A table would be helpful to summarize the results presented for each case study

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on the fraction of trajectories entering the Arctic.

We have carefully considered this comment, but we believe such a table would only make the manuscript longer. The table would have only three pieces of information for each of three geographic regionsâĂŤonly nine cells. Even with a table, the results would continue to have to be discussed and compared in the text. Therefore we hope it is acceptable not to include tables of this type.

13. The Summary and Conclusions section should be condensed by 50% and the main conclusions more clearly brought forward.

We have shortened this section to the best of our ability.

Detailed Comments

Abstract

1. Pg. 5436, L. 13: Trajectories can be calculated but not released, air parcels can be released and traced. Use consistent wording throughout the manuscript.

Of course, your wording is correct and more precise than ours. “Released” means the starting point of the trajectory calculations. We have changed the wording in the abstract and the main text, often substituting “initiated”

2. L. 14: historically significant: not clear what you mean, rephrase. This applies at all instances of this term throughout the manuscript.

We have changed the wording throughout the text. We mean “indicated by literature”.

3. L. 12-20 can be shortened

These lines have been shortened.

4. L. 20: the term climatology, even though in quotation marks, does not apply for the mean characteristics over a one-month period.

This has been changedâĂŤsee comment 8 above.

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Introduction

5. Pg. 5437, L. 7: citing Stohl (2006) and Law and Stohl (2007) in that location sounds as if those papers would argue for seeing the Arctic as a clean location - rephrase!

We did not mean to imply that these studies thought the Arctic currently was clean. However, Stohl (2006) did state that the Arctic used to be considered clean. We have removed the other reference and rephrased the sentence to make this point clear.

6. There are also pollution sources well within the Arctic from natural gas and oil extraction and other heavy industry.

We have added this statement.

7. L. 20: citing Stohl (2006) and Warneke et al (2009) in this context is wrong, these studies actually do note the importance of non-anthropogenic pollution for Arctic air pollution

We have removed these references.

8. Pg. 5438, L. 11: cold potential temperature -> should be low, potential temperature is a measure of energy

We respectfully disagree. Our reasoning is as follows. Because potential temperature is considered a temperature (see AMS Glossary of Meteorology), we believe that the words “high” and “low” should refer only to altitude wherever possible. This avoids any possibility of confusion. Thus, “cold” is the best way to describe a temperature.

9. L. 21: Stohl and Law (2006) does not seem to be peer-reviewed literature, is it necessary to cite this?

It is not absolutely necessary. We have removed it.

10. L. 23: "lifted over the Arctic front": not clear what you mean, rephrase.

We have rephrased.

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11. Pg. 5439, L. 10: could remove Jacob reference here

Done.

Data and Methodology

12. Pg. 5440, L. 20: motivate using the limited nests, and using WRF over GFS in the first place

See comment 1 above. We could obtain higher spatial (45 km) and temporal resolution (we used hourly) from WRF. GFS data normally are not available at 1 h resolution, although they might exist somewhere in the NCEP archives. If so, it would have been difficult to acquire them. GFS data would not have utilized the higher resolution nesting which we thought was important in the areas of greatest emissions. WRF is a very widely used model. We have added information about this to the revision.

13. Pg. 5441, L. 4: Trajectories -> 3D kinematic trajectories

This has been rephrased to include your point.

14. L. 8: Mention specifically what advantages particle dispersion models have, and why you think trajectories are more appropriate for your study.

See our response to item 4 of reviewer 1

15. L. 15: clarify how you specify starting locations from the emissions inventory. How is the emissions inventory different to e.g. EDGAR?

The description of starting locations has been revised.

See major comment 3 above about the emissions inventory.

16. Fig 2: How were release locations specified? In Fig. 2e it appears that France has been left out from the analysis, why? Also, why have Turkey and the Iraq been included but not the European part of Russia and the Ukraine?

We have elaborated on our release specifications in the Methodology section (Section

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2.2). We did not release from every location having anthropogenic emissions, but only from those having relatively strong emissions.

17. L. 16: "release" -> starting

This paragraph has been rephrased to include your terminology.

18. L. 20: at what level where the trajectories released?

All forward trajectories were initiated at the surface. This is emphasized in Section 2.2 of the revision

19. L. 26: What distance did the aircraft cover during that time interval?

It all depends on the altitude of the aircraft. The DC-8 typically flew at a speed of $\sim$400 kt in the upper troposphere and $\sim$250 kt in the lower troposphere. We now cite distances for these two aircraft speeds.

20. Fig. 4, 9, 13: Maps in panels b and c should be rotated to the same center longitude

These panels have been reoriented as requested.

21. Pg. 5444, L. 23: "hot spots" -> secondary maxima

This has been rephrased.

22. Pg. 5445, L. 5: combine this information into a table for all regions

Our concern is that such a table would simply make the manuscript longer. The table would have only three pieces of information for each of three geographic regionsâĂŤonly nine cells. Even with a table, the results would continue to have to be discussed and compared in the text.

23. L. 18: standard techniques: please include more information here on how this has been done.

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We have taken details from the web site that was originally cited and inserted them into the text.

24. Pg. 5446, L. 2: mention the sources of biomass burning here

We have added the references cited previously that state that most of the biomass emissions are from eastern Asia.

25. Pg. 5447, L. 27: formulate more carefully here, it is likely/probably a source in China, but I don't see why you could rule out for instance a source in Japan.

We have studied Asian outflow extensively during the past decade, and we have been able to show that there is a specific fingerprint for air of Chinese origin, and that this is distinct from air of Japanese or Korean origin. Unlike the Chinese fingerprint of H-1211/OCS/1,2-DCE/CH3Cl (Barletta et al., 2009), we have previously observed elevated levels of CH3Br in air of Japanese/Korean origin (Blake et al., 2003). Therefore we are able to conclude that the observed air is of Chinese origin, and is not from Japan. To address the reviewer's comment we have added the following text on Line 27: "Note that air from other regions of Asia such as Japan or Korea have their own chemically distinct signatures (Blake et al., 2003), and we are able to specifically attribute the leg 5 samples to source influences from China"..

26. Pg. 5448, L. 8: constant pressure analysis: not clear what you mean, sea level pressure?

Although we examined various meteorological parameters on many constant pressure surfaces (e.g., 850, 700, 500 and 300 hPa), we only present results for sea level pressure. We have changed the sentence to reflect this.

27. L. 11-15: rewrite, it is confusing to describe the backward calculation as a "release from the aircraft"

This has been rephrased.

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28. L. 20: how does the uncertainty of trajectory calculations, that can amount to thousands of km after a few days, affect your interpretation of this transport pattern?

We are very aware of the uncertainty in trajectories, and we cite several references on this topic. We have strived to incorporate these uncertainties in our discussions. We made another pass through the text to make sure that we only mention major transport features that hopefully are not overly influenced by trajectory uncertainty.

29. Pg. 5449, L. 1: Branches should be labeled or otherwise made distinguishable in Fig. 6

We have labeled the figure as requested and changed its caption.

30. L. 28 onward: place into method section once for all cases, add table for results.

We have moved this material to new Section 2.3 as requested. However, there still must be some mention of the procedure in the various case studies. However, those discussions now can be much shorter.

31. Pg. 5450, L. 23 onward: remove summary paragraphs here and in the other case studies, they contain unnecessary repetition.

We respectfully disagree with the reviewer's suggestion. The text has just finished a detailed discussion of the East Asian region. It is good practice to write a brief summary (8 lines here) before moving to the next geographic region which has somewhat different characteristics. We believe the summaries will assist most readers and are a matter of personal preference. The other two reviewers did not request their removal, and we hope it will be acceptable that we have retained each of the summaries.

32. Pg. 5451, L. 7: "the distribution of Arctic arrivals: : :" unclear, rephrase

This has been rephrased. We meant the locations whose trajectories reach the Arctic mostly are in the northern half of the domain.

33. Pg. 5452, L. 16: list the species here

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We have inserted the species into the text as requested.

34. Pg. 5454, L. 9: again, how reliable is it to follow individual trajectories over 10 days given the considerable errors individual trajectories may have?

Please see our response to comment 28.

35. Pg. 5455, L. 11-18: delete summary section

See comment 31.

36. Pg. 5456, L. 5: releases were made in Turkey and the middle east, it appears that this is rather responsible for the apparent transport maximum.

We have modified the wording to add countries and locations east of the domain. This is still a valid pathway.

37. L. 7: "middle and upper levels": are not all trajectories released at the surface? Unclear.

Yes, all trajectories were initiated at the surface, but during their 15 day transit, many ascend to much higher altitudes. We have rephrased this.

38. L. 17: "Thus, considering the combination: : :" Not clear what is your point here. Asia, America and Europe span a range of longitudes, and so do the entry regions. This should not be surprising.

It is not surprising. The sentence is just a simple way of summing up the findings of the three source regionsâĂŤthe pollution comes from many locations. We have modified this sentence to make our point clearer.

39. L. 25: To what extent is this due to the more northerly starting locations in Europe compared to the other regions?

This is an important point that we have added to the revision.

40. Pg. 5458, L. 16: remove summary section

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See comment 31.

41. Pg. 5459, L. 2: It appears as an over-interpretation to rely on the accuracy of trajectories calculated several days backward in time to the degree needed for your interpretation. In particular in the vicinity of the boundary layer the concept of conserved air parcels becomes increasingly questionable due to turbulent mixing. To fully exclude a biomass burning influence in your airmass you would need the information from a specific tracer, such as levoglucosan.

We respectfully disagree that we have over-interpreted the trajectories, because the trajectory results supplement the results of the chemical data throughout the paper. The chemistry proves anthropogenic origins and in some cases ties the pollution directly to a source region. If we can show through chemistry that we have anthropogenic influences and our trajectories are released from known areas of enhanced anthropogenic emissions and reach the aircraft, then we can make a strong conclusion that we sampled anthropogenic emission. Levoglucosan was not measured during this mission, but other long-lived biomass burning tracers such as HCN and CH3CN were. To address the reviewer's comment we have added the following text on Line 12: "Consistent with this, a biomass burning signature is also not evident based on the measured mixing ratios of the biomass burning tracers HCN and CH3CN (not shown). Thus, the brief fire area boundary layer encounters may be an artifact of trajectory uncertainty."

42. Pg. 5460, L. 11-20: remove/condense summary section

See response to comment 41.

43. L. 22: The proximity of the "entry location" to Asia makes it very likely that this airmass was indeed a mix of air masses, that may have included European pollution, but unlikely undiluted (by, for instance, Asian pollution sources, as encountered earlier in the flight) over such a long travel distance.

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This scenario certainly is possible. However, we did not detect a known chemical signature from Asia, and a specific European signature is not documented in the literature. We do not claim that the mixture is purely European, only that it is purely anthropogenic and likely has at least a partial European origin. We have modified the sentences accordingly.

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