Peer Review File

Mysterious air pollution in south China linked to volcanic emissions from the Philippines

Corresponding Author: Dr Changqing Lin

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Decision Letter:

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Dear Dr Lin,

Your manuscript titled "Mysterious air pollution in south China is linked to volcanic emissions in the Philippines" has now been seen by 3 reviewers, whose comments are appended below. You will see that they find your work of some potential interest. However, they have raised quite substantial concerns around the robustness of your proposed index, that must be addressed. In light of these comments, we cannot accept the manuscript for publication in its current form, but would be interested in considering a revised version that fully addresses these serious concerns.

Specifically, we require that you:
-rigorously demonstrate that air pollution in South China originates from the Taal volcano
-test your index against other volcanic events to examine its robustness to different conditions.

We hope you will find the reviewers' comments useful as you decide how to proceed. Should additional work allow you to address these criticisms, we would be happy to look at a substantially revised manuscript. If you choose to take up this option, please either highlight all changes in the manuscript text file, or provide a list of the changes to the manuscript with your responses to the reviewers.

When resubmitting, please provide a point-by-point response to the reviewers' comments. Please submit your responses as a separate file, distinct from your cover letter where you can add responses to the Editors' comments that you do not want to be made available to the reviewers. Word files are preferred. We recommend that any figures, tables or graphs that are included in the response to reviewers are also included in the main article or Supplementary Information.

Please bear in mind that we will be reluctant to approach the reviewers again in the absence of substantial revisions.

If the revision process takes significantly longer than three months, we will be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

We are committed to providing a fair and constructive peer-review process. Please do not hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the reviewers' comments with a list of your changes to the manuscript text (which should be in a separate document to any cover letter), a tracked-changes version of the manuscript (as a PDF file) and any completed checklist:

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Please do not hesitate to contact us if you have any questions or would like to discuss the required revisions further. Thank you for the opportunity to review your work.

Best regards,

Alice Drinkwater, PhD
Associate Editor
Communications Earth & Environment
@CommsEarth

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(https://www.nature.com/documents/commsj-phys-style-formatting-guide-accept.pdf).

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

The paper presents evidence of Taal volcano transport to southern China as measured from Hong Kong and stations in PRD. It also formulated a simple index to identify Taal as the source of local/regional pollutants. The topic is interesting and the manuscript is well-written. I do have some questions/comments to clarify some of the points presented before the manuscript is recommended for publication.

- consider this rewording: "Mysterious air pollution in south China linked to volcanic emissions from the Philippines"

- May I suggest that from the abstract, indicate or make it clear that measurements are mostly done in HK, instead of just only stating South China (first)? For instance, "A mysterious air pollution event occurred in south China" + ,as measured from HK, ..., same in the introduction. So that there is no confusion on where the study was done from the 1st line.

- Abstract: perhaps remove the "with concentrations often below detection limits" in the abstract and just state in the main body.

- Figure 1, Please add a unit label to the color scale, or at least in the figure caption

- Line 76: it would be helpful to indicate what satellite

- Line 126: can the authors contextualize the value of 73.1 (since it's from a self-defined index), what does this value mean? what value is considered high/low?

- Line 135: what is meant by variations in wind pattern? I understand that volcanic emission varies, that's a given. For wind, do you mean to say that even if wind measurements are southerly in HK, winds in the South China Sea may not transport Taal emissions northward (assuming there's emission)? So, does this mean that the CWI should also include a measure of how easterly and (not just how) southerly the winds in Taal and the South China Sea, respectively, are? Why or why not?

- On CWI, were the authors able to try percentile values lower than 99? such as 95? I was wondering if the 99th percentile is too strict. Does the April 5-6 measurement of P in fig 3a exceed the 99th percentile?

- In relation to the previous comment, it seems like the appearance of P is essentially the basis for volcanic emission transport, I base this on the conclusion that April 5 was tagged as volcanic even if there were no S values, how was the CWI calculated for that case? and were there any elevated P values in the past 3 years occurring with southerly winds that were not related to Taal?

- In summary, my question is mainly about the robustness of CWI, since there's only a 2-week period on which the equation is based, and no other cases were found in the previous 3 years, how sure are the authors that this will work in most (or all) of the instances where volcanic emissions are transported? I think the paper should drive more on this.

Reviewer #2 (Remarks to the Author):

Review report for manuscript titled "Mysterious air pollution in south China is linked to volcanic emissions in the Philippines" by Dr Lin and colleagues,

Question: What are the major claims of the paper?

Answer: The pollution in south China is a result of the eruption of the Taal Volcano in the Philippines.

Question: Are these claims novel, and will they be of interest to others in the community? If the conclusions are not original, it would be helpful if you could provide relevant references.

Answer: Akilan et al., 2019 and Akilan et al., 2023 observed nearly identical results in an alternative area, as evidenced by their research conducted in Antarctica and India.

Question: Is the work convincing, and if not, what further evidence would be required to strengthen the conclusions?

Answer: Additional evidence is unnecessary at this time. Nevertheless, the authors have the opportunity to explore historical events that may have taken place in the same location at the earlier time.

Question: On a more subjective note, do you feel that the paper will influence thinking in the field in terms of either conceptual understanding or technological capability? Please feel free to raise any further questions and concerns about the paper.

Answer: Yes
Suggestions:
1. Further comprehension can be achieved by conducting studies using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model.
2. A Trend line could be inserted into the illustration to aid the readers in better understanding.
3. Exhibit transboundary.

Reviewer #3 (Remarks to the Author):

The research article titled “Mysterious air pollution in south China is linked to volcanic emissions in the Philippines” is an interesting study.
However, the following corrections should be considered before publication:
Line 31-32, authors should add data in support of their statement.
What is the height of the observed air pollutants i.e PM2.5.
Figure3: Please show measurements of pollutants from February to May.
Providing a more precise explanation of the study's novelty is recommended. What specific contributions does this research make to the existing literature.
The introduction could be improved by incorporating more relevant and recent references.

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Version 1:

Decision Letter:

** Please ensure you delete the link to your author home page in this e-mail if you wish to forward it to your coauthors **

Dear Dr Lin,

Your manuscript titled "Mysterious air pollution in south China linked to volcanic emissions from the Philippines" has now been seen by our reviewers, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment.

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewers. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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Please review our specific editorial comments and requests regarding your manuscript in the attached "Editorial Requests Table".

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We hope to hear from you within two weeks; please let us know if you need more time.

Best regards,

Alice Drinkwater, PhD
Associate Editor

Communications Earth & Environment
@CommsEarth

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The manuscript is much improved in this version. I have a few minor comments though.

1. Perhaps in the first mention of CTM, indicate that it is based on WRF-CMAQ model so the readers will have an idea that there's chemistry in the model.

2. The backward trajectory for 1 April is off the south of Taal, may I suggest that the authors use an ensemble of back trajectories, also with varying heights from near surface to 500m since surface concentrations were also affected, just to reduce the uncertainty of using a single trajectory. This should be easy to redo.

3. Can the authors add some more information about the lidar in Fig. S5, can just be included in the figure caption. Information such as general type of lidar, wavelength, resolution, etc.

4. Line 205: Don't start a paragraph with "however". Rephrase the first sentence.

Also, when submitting a revision, include a clean version and a tracked version.

Reviewer #2 (Remarks to the Author):

The manuscript is acceptable in its current form

Re: COMMSENV-24-1846-T (*Mysterious air pollution in south China is linked to volcanic emissions in the Philippines*)

Dear Editor and Reviewers,

We are grateful to the editor and reviewers for their helpful comments. Modifications have been made in response to these comments. Below is a point-by-point response to each comment. In the responses, text in black represents the reviewers' comments, while text in red represents our responses. A change-tracked version of the manuscript is also uploaded for your reference. We hope that you and the referees find the changes satisfactory, and we look forward to hearing from you soon.

Dr. Changqing LIN, E-mail: cqlin@ust.hk

Division of Environment and Sustainability

The Hong Kong University of Science and Technology

December 4, 2024

Reviewer #1 (Remarks to the Author):

The paper presents evidence of Taal volcano transport to southern China as measured from Hong Kong and stations in PRD. It also formulated a simple index to identify Taal as the source of local/regional pollutants. The topic is interesting and the manuscript is well-written. I do have some questions/comments to clarify some of the points presented before the manuscript is recommended for publication.

- consider this rewording: "Mysterious air pollution in south China linked to volcanic emissions from the Philippines"

Response: We greatly appreciate your comment. We revise the title according to your comment.

- May I suggest that from the abstract, indicate or make it clear that measurements are mostly done in HK, instead of just only stating South China (first)? For instance, "A mysterious air pollution event occurred in south China" + ,as measured from HK, ..., same in the introduction. So that there is no confusion on where the study was done from the 1st line.

Response: We greatly appreciate your comments. In the Abstract, we specify that Hong Kong is the city for obtaining chemical measurement data (lines 23-29):

> "In this study, we identified the source of this event using a multi-scale data system that combines large-scale monitoring and modeling data with advanced measurements of aerosol chemical compositions in Hong Kong. Large-scale satellite measurements and chemical transport modeling detected the transboundary transport of a volcanic plume originating from the Taal volcano in the Philippines. Unusually high levels of sulfate (reaching 75% of fine particulate matter) and phosphorus (up to 93 ng/m^3) in Hong Kong supported this finding."

In the Introduction Section, we highlight Hong Kong as a key study city (lines 92-97):

> "In this study, we aim to evaluate the impacts of volcanic emissions on air quality by using a multi-scale data system that combines large-scale data (e.g., satellite monitoring and CTM simulations) with advanced monitoring of aerosol chemical compositions at specific stations in Hong Kong. The real-time multi-scale data were used to identify the potential

sources and long-range transport mechanisms responsible for the mysterious air pollution event that occurred in south China, particularly in Hong Kong, on April 1-2, 2024."

- Abstract: perhaps remove the "with concentrations often below detection limits" in the abstract and just state in the main body.

Response: Thank you very much for your valuable comments. We remove this statement in the Abstract.

- Figure 1, Please add a unit label to the color scale, or at least in the figure caption

Response: Thank you very much for your comments. We added the unit for $PM_{2.5}$ (unit: μg/m³) in the caption of Figure 1.

"Spatial distributions of $PM_{2.5}$ concentration (unit: μg/m³) in Hong Kong and its adjacent Pearl River Delta (PRD) region at 10 am, 2 pm, 6 pm, and 10 pm on April 1, 2024 (c)."

- Line 76: it would be helpful to indicate what satellite

Response: Thank you very much for your comments. We clarify the satellite's name for this statement (lines 103-106):

"According to the vertical column density of SO_2 from GEMS measurements and the true-color image from MODIS aboard Aqua on March 30, 2024 (Supplementary Fig. S3a, b), the Taal volcano released a large amount of SO_2 and ash into the atmosphere, which was then transported westward over the SCS."

- Line 126: can the authors contextualize the value of 73.1 (since it's from a self-defined index), what does this value mean? what value is considered high/low?

Response: Thank you very much for your comments. We clarify the meaning of the CWI value as defined in our framework (lines 161-163)"

> "Details of the CWI are shown in the Method Section. A positive CWI value above one suggests that it is very likely the pollution in Hong Kong is associated with the volcanic emissions from the Philippines."

- Line 135: what is meant by variations in wind pattern? I understand that volcanic emission varies, that's a given. For wind, do you mean to say that even if wind measurements are southerly in HK, winds in the South China Sea may not transport Taal emissions northward (assuming there's emission)? So, does this mean that the CWI should also include a measure of how easterly and (not just how) southerly the winds in Taal and the South China Sea, respectively, are? Why or why not?

Response: Thank you very much for your valuable comments. The suitable wind conditions for transboundary transport include at least easterly winds at Taal and southerly winds in Hong Kong. These are the basic requirements for the wind pattern to transport pollutants from the Taal volcano to Hong Kong. The frequency of these wind conditions varies over time. Therefore, we assess the occurrence frequency of these conditions. We revise the sentence to clarify the meaning of "variation in wind pattern" (lines 177-179):

> "This can be explained by the variations in volcanic activity and occurrence frequency of the suitable wind patterns for transboundary transport."

We agree with the reviewer that wind conditions in Hong Kong, the Philippines, and even the South China Sea can affect the transport of air pollutants. In this study, the development of the CWI focuses on its application in a city impacted by volcanic emissions. Therefore, the index relies solely on local data from this city and does not require data from other regions. The use of local data in the CWI facilitates its application in cities where chemical and meteorological measurements are available. We clarify the rationale for using only local data in the CWI (lines 198-207):

> "Wind conditions in Hong Kong, the Philippines, and even the SCS can affect the transport of air pollutants. In this study, the development of the CWI focuses on its application in a city impacted by volcanic emissions. Therefore, the index relies solely on local data from this city and does not require data from other regions. The development and utilization of the CWI, which integrates local chemical and wind data, provide a robust approach for specific regions to identify the influence of the transported volcanic plumes. The use of

> local data in the CWI facilitates its application in cities where chemical and meteorological measurements are available. This index is particularly useful for people residing in areas at risk of volcanic activities, helping to protect their public health. The results of this study support emergency preparedness and help minimize the adverse impacts on the population."

- On CWI, were the authors able to try percentile values lower than 99? such as 95? I was wondering if the 99th percentile is too strict. Does the April 5-6 measurement of P in fig 3a exceed the 99th percentile?

Response: We greatly appreciate your comments.

(1) As shown in the Method Section, the 99th percentile of S and P concentrations under the southerly winds were estimated to be 4.6 μg/m^3 and 5.1 ng/m^3, respectively. During all four pollution episodes, we found unusually high P levels (peaking at 93.1 ng/m^3, 30.7 ng/m^3, 42.3 ng/m^3, and 38.8 ng/m^3), which significantly exceeded its 99th percentile of 5.1 ng/m^3. We clarify the concentration level of P in lines 167-171:

> "During these four pollution episodes, we found unusually high levels of S (peaking at 18.4 μg/m^3, 13.6 μg/m^3, 11.0 μg/m^3, and 6.8 μg/m^3), which significantly exceeded its 99th percentile of 4.6 μg/m^3. Additionally, we found unusually high P levels (peaking at 93.1 ng/m^3, 30.7 ng/m^3, 42.3 ng/m^3, and 38.8 ng/m^3), which also significantly exceeded its 99th percentile of 5.1 ng/m^3."

(2) In fact, we tested the 95th percentile values. Since the P concentration often falls below detection limits (recorded as zero), the 95th percentile for P concentration is identified as zero. Given this situation, we used the 99th percentile, instead of the 95th percentile, as the threshold for P concentration.

- In relation to the previous comment, it seems like the appearance of P is essentially the basis for volcanic emission transport, I base this on the conclusion that April 5 was tagged as volcanic even if there were no S values, how was the CWI calculated for that case? and were there any elevated P values in the past 3 years occurring with southerly winds that were not related to Taal? **Response:** We greatly appreciate your comments. This study used sulfate (SO_4^{2-}) concentrations measured by MARGA and sulfur (S) concentrations measured by XRF spectrometers. We apologize for the confusion in the figure, which plots sulfate and P concentrations. Occasionally, MARGA measurements of sulfate were unavailable due to equipment issues. In this study, the CWI value is calculated based on the S and P concentrations measured by XRF spectrometers. To avoid confusion, we have replotted Figure 3 to display the time series of S and P concentrations.

In addition to the case during April 1-2, we identified three additional air pollution events from April to June 2024 (e.g., April 5, April 15-16, and June 21-22) that could be attributed to ongoing emissions from the Taal volcano. During these four air pollution episodes, we found unusually high levels of both S and P concentrations. Results are shown in lines 165-173:

> "Additionally, the application of this index identified three additional air pollution events from April to June 2024 (e.g., April 5, April 15-16, and June 21-22) that could be attributed to ongoing emissions from the Taal volcano. During these four pollution episodes, we found unusually high levels of S (peaking at 18.4 μg/m^3, 13.6 μg/m^3, 11.0 μg/m^3, and 6.8 μg/m^3), which significantly exceeded its 99th percentile of 4.6 μg/m^3. Additionally, we found unusually high P levels (peaking at 93.1 ng/m^3, 30.7 ng/m^3, 42.3 ng/m^3, and 38.8 ng/m^3), which also significantly exceeded its 99th percentile of 5.1 ng/m^3. During all these pollution episodes, increases in P levels coincided with elevated S concentrations, and the dominant winds in Hong Kong were southerly."

- In summary, my question is mainly about the robustness of CWI, since there's only a 2-week period on which the equation is based, and no other cases were found in the previous 3 years, how sure are the authors that this will work in most (or all) of the instances where volcanic emissions are transported? I think the paper should drive more on this.

Response: Thank you very much for your valuable comments.

(1) We extend the duration of the study period. The application of the CWI index identified four air pollution events from April to June 2024 (e.g., April 1-2, April 5, April 15-16, and June 21-22) that could be attributed to ongoing emissions from the Taal volcano. During these four air pollution episodes, we found unusually high levels of S and P. Increases in P levels coincided with elevated S concentrations, and the dominant winds in Hong Kong were southerly. This evidence from

meteorological conditions and chemical measurements supports the transboundary transport of volcanic pollutants. New results are shown in Figure 3 and in lines 164-174:

> "The results show that the CWI reached a maximum of 73.1 at 6 pm on April 1 (Fig. 3b). Additionally, the application of this index identified three additional air pollution events from April to June 2024 (e.g., April 5, April 15-16, and June 21-22) that could be attributed to ongoing emissions from the Taal volcano. During these four pollution episodes, we found unusually high levels of S (peaking at 18.4 $\mu g/m^3$, 13.6 $\mu g/m^3$, 11.0 $\mu g/m^3$, and 6.8 $\mu g/m^3$), which significantly exceeded its 99th percentile of 4.6 $\mu g/m^3$. Additionally, we found unusually high P levels (peaking at 93.1 ng/m^3, 30.7 ng/m^3, 42.3 ng/m^3, and 38.8 ng/m^3), which also significantly exceeded its 99th percentile of 5.1 ng/m^3. During all these pollution episodes, increases in P levels coincided with elevated S concentrations, and the dominant winds in Hong Kong were southerly. This evidence from meteorological conditions and chemical measurements supports the transboundary transport of volcanic pollutants."

(2) The application of CWI did not detect additional significant air pollution cases related to volcanic emissions in Hong Kong during the previous three years. This can be explained by the variations in volcanic activity and occurrence frequency of the suitable wind patterns for transboundary transport. For instance, the suitable wind conditions occur less frequently in winter due to the dominant northerly winds that are associated with the winter monsoon. In contrast, the occurrence frequency of suitable wind conditions for transboundary transport greatly increased to 50.9% in April 2024. The increased occurrence of suitable wind conditions has led to the noticeable increase in air pollution episodes in Hong Kong that are related to the Taal volcano emissions in April 2024. Therefore, the varying impacts of volcanic emissions on air quality in south China, both in April 2024 and over the past three years, can be explained by our CWI system. These results support the robustness of the CWI system. We clarify the reasons for the varying impacts of volcanic emissions in lines 176-189:

> "However, the application of CWI did not detect any additional significant air pollution cases related to volcanic emissions in Hong Kong during the previous three years. This can be explained by the variations in volcanic activity and occurrence frequency of the suitable wind patterns for transboundary transport. According to the emission data (Supplementary

Fig. S12a), the volcano was less active during April 2022 and September 2023. The suitable wind conditions for transboundary transport include easterly winds at Taal and southerly winds in Hong Kong. These suitable wind conditions occurred less frequently in winter due to the dominant northerly winds that are associated with the winter monsoon (Supplementary Fig. S12b). In contrast, the occurrence frequency of suitable wind conditions for transboundary transport greatly increased to 50.9% in April 2024. The increased occurrence of suitable wind conditions has led to the noticeable increase in air pollution episodes in Hong Kong that are related to the Taal volcano emissions in April 2024. Therefore, the varying impacts of volcanic emissions on air quality in south China, both in April 2024 and over the past three years, can be explained by our CWI system. These results support the robustness of the CWI system."

Reviewer #2 (Remarks to the Author):

Review report for manuscript titled "Mysterious air pollution in south China is linked to volcanic emissions in the Philippines" by Dr Lin and colleagues,

Question: What are the major claims of the paper?

Answer: The pollution in south China is a result of the eruption of the Taal Volcano in the Philippines.

Question: Are these claims novel, and will they be of interest to others in the community? If the conclusions are not original, it would be helpful if you could provide relevant references.

Answer: Akilan et al., 2019 and Akilan et al., 2023 observed nearly identical results in an alternative area, as evidenced by their research conducted in Antarctica and India.

Response: We greatly appreciate your comments. We have thoroughly rewritten the Introduction to incorporate more relevant references and clarify our novelty (lines 64-100):

> "Accurately tracking pollution sources for such events requires large-scale data from diverse domains. Akilan et al. (2019, 2023) used GPS data to demonstrate the impacts of volcanic eruptions around the Antarctic continent and in India[9,10]. With improvements in computational capability, state-of-the-art chemical transport models (CTMs) have become advanced tools increasingly used to reveal the transport of air pollutants from volcanoes[11].

Based on a CTM model, Filonchyk et al. (2022) explored the transport of air pollutants from the volcanic eruption in the Canary Islands[12]. Inness et al. (2022) enhanced the capability of another CTM to forecast pollutant transport from the 2019 Raikoke eruption by assimilating SO_2 column and layer height data[13]. Additionally, large-scale satellite measurements have proven useful for revealing emissions and the long-range transport of air pollutants from volcanoes[12,14]. For instance, based on satellite measurements, Milford et al. (2023) evaluated the impact of the 2021 La Palma volcanic eruption on air quality[15].

Most of the aforementioned studies used large-scale data to evaluate the impacts of volcanic eruptions by focusing on pollutants that are regularly monitored, such as $PM_{2.5}$ and SO_2. These large-scale analyses may face two challenges. First, the concentration of $PM_{2.5}$ is influenced by various anthropogenic and natural sources. Therefore, an increase in $PM_{2.5}$ concentration may not necessarily be related to volcanic emissions, complicating the assessment. Second, the concentration of SO_2 may rapidly decrease during long-range transport as it converts to sulfate, which is a component of particulate matter.

Analysis of the concentration of tracer components in aerosols provides direct evidence of the impacts of volcanic emissions[16]. It is widely recognized that volcanoes are significant contributors to the sulfur (S) budget and that volcanic activities lead to increased aerosol S concentrations[19]. Additionally, volcanic ash contains high levels of phosphorus (P), which can promote the growth of vegetation in areas affected by ash deposition[17,18]. During a volcanic event in Hawaii, elevated concentrations of aerosol S and P were detected at the Hawaiian Volcano Observatory[20]. Therefore, detailed measurements of aerosol chemical composition, even at specific locations, can significantly aid in confirming pollution sources during events influenced by volcanoes.

In this study, we aim to evaluate the impacts of volcanic emissions on air quality by using a multi-scale data system that combines large-scale data (e.g., satellite monitoring and CTM simulations) with advanced monitoring of aerosol chemical compositions at specific stations in Hong Kong. The real-time multi-scale data were used to identify the potential sources and long-range transport mechanisms responsible for the mysterious air pollution event that occurred in south China, particularly in Hong Kong, on April 1-2, 2024. This multi-scale approach exemplifies comprehensive analyses that accurately attribute air pollution to unusual emission sources, such as volcanoes. Additionally, it enhances emergency preparedness and reduces health risks in regions vulnerable to volcanic events."

Question: Is the work convincing, and if not, what further evidence would be required to strengthen the conclusions?

Answer: Additional evidence is unnecessary at this time. Nevertheless, the authors have the opportunity to explore historical events that may have taken place in the same location at the earlier time.

Response: Thank you very much for your valuable comments.

(1) We replot Figure 3 to extend our study period. We found more air pollution cases that could be attributed to ongoing emissions from the Taal volcano. Results are shown in lines 165-174:

> "Additionally, the application of this index identified three additional air pollution events from April to June 2024 (e.g., April 5, April 15-16, and June 21-22) that could be attributed to ongoing emissions from the Taal volcano. During these four pollution episodes, we found unusually high levels of S (peaking at 18.4 μg/m^3, 13.6 μg/m^3, 11.0 μg/m^3, and 6.8 μg/m^3), which significantly exceeded its 99th percentile of 4.6 μg/m^3. Additionally, we found unusually high P levels (peaking at 93.1 ng/m^3, 30.7 ng/m^3, 42.3 ng/m^3, and 38.8 ng/m^3), which also significantly exceeded its 99th percentile of 5.1 ng/m^3. During all these pollution episodes, increases in P levels coincided with elevated S concentrations, and the dominant winds in Hong Kong were southerly. This evidence from meteorological conditions and chemical measurements supports the transboundary transport of volcanic pollutants."

(2) Additionally, we re-examine air pollution cases in Hong Kong during the past three years. However, we did not detect additional significant air pollution cases related to volcanic emissions. This can be explained by the variations in volcanic activity and occurrence frequency of the suitable wind patterns for transboundary transport. Results are shown in lines 176-189:

> "However, the application of CWI did not detect any additional significant air pollution cases related to volcanic emissions in Hong Kong during the previous three years. This can be explained by the variations in volcanic activity and occurrence frequency of the suitable

wind patterns for transboundary transport. According to the emission data (Supplementary Fig. S12a), the volcano was less active during April 2022 and September 2023. The suitable wind conditions for transboundary transport include easterly winds at Taal and southerly winds in Hong Kong. These suitable wind conditions occurred less frequently in winter due to the dominant northerly winds that are associated with the winter monsoon (Supplementary Fig. S12b). In contrast, the occurrence frequency of suitable wind conditions for transboundary transport greatly increased to 50.9% in April 2024. The increased occurrence of suitable wind conditions has led to the noticeable increase in air pollution episodes in Hong Kong that are related to the Taal volcano emissions in April 2024. Therefore, the varying impacts of volcanic emissions on air quality in south China, both in April 2024 and over the past three years, can be explained by our CWI system. These results support the robustness of the CWI system."

Question: On a more subjective note, do you feel that the paper will influence thinking in the field in terms of either conceptual understanding or technological capability? Please feel free to raise any further questions and concerns about the paper.

Answer: Yes

Suggestions:

1. Further comprehension can be achieved by conducting studies using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model.

Response: Thank you very much for your valuable comments. We add analyses using the HYSPLIT model. Results are shown in supplementary Fig. S4 and in lines 114-116:

"Furthermore, the trajectory of volcanic pollutants simulated using the HYSPLIT model shows similar patterns of transboundary transport from the Philippines to Hong Kong (Supplementary Fig. S4)."

2. A Trend line could be inserted into the illustration to aid the readers in better understanding.

Response: Thank you very much for your valuable comments. In addition to satellite monitoring and model simulations, we added analyses using the HYSPLIT model to further emphasize the trend line of volcanic pollutants from the Philippines to Hong Kong. Results are shown in supplementary Fig. S4.

3. Exhibit transboundary.

Response: We greatly appreciate your comments. The transboundary transport of volcanic pollutants is presented in the first part of the Results section. First, the vertical column density of SO_2 and true-color images from satellite measurements were used to track the transport of air pollutants. Second, CTM simulations of sulfate concentration confirmed this transboundary transport. Third, we included analyses using the HYSPLIT model to further emphasize the transboundary transport of volcanic pollutants from the Philippines to Hong Kong. Results are shown in lines 103-116.

> "According to the vertical column density of SO_2 from GEMS measurements and the true-color image from MODIS aboard Aqua on March 30, 2024 (Supplementary Fig. S3a, b), the Taal volcano released a large amount of SO_2 and ash into the atmosphere, which was then transported westward over the SCS. The CTM simulations of sulfate (SO_4^{2-}) concentration (Supplementary Fig. S3c) showed a similar track of the volcanic plume. During the pollution episode on April 1, 2024, the satellite measurements show that the volcano-generated gas and aerosol were initially carried westward, then turned northward and eventually reached south China with the predominantly southerly winds over the SCS (Fig. 2a, b). During the transport of volcanic pollutants, SO_2 was converted to sulfuric acid, rapidly condensing to form sulfate aerosols, leading to secondary air pollution issues[21]. The CTM simulations of the SO_4^{2-} concentration show a similar track of the volcanic plume and confirm the transport of the volcanic aerosol from the Taal volcano to south China (Fig. 2c). Furthermore, the trajectory of volcanic pollutants simulated using the HYSPLIT model shows similar patterns of transboundary transport from the Philippines to Hong Kong (Supplementary Fig. S4)."

Reviewer #3 (Remarks to the Author):

The research article titled "Mysterious air pollution in south China is linked to volcanic emissions in the Philippines" is an interesting study. However, the following corrections should be considered before publication:

¬ Line 31-32, authors should add data in support of their statement.

Response: We greatly appreciate your comments. We add Fig. S1 to the supplementary file to illustrate the dependence of $PM_{2.5}$ concentration on wind conditions. Results are presented in lines 35-37:

> "In south China, concentrations of fine particulate matter ($PM_{2.5}$) are generally low under southerly winds, which bring clean air from the South China Sea (SCS)[1,2]. This is illustrated by pollution rose using measurement data from Hong Kong (Supplementary Fig. S1)."

¬ What is the height of the observed air pollutants i.e PM2.5.

Response: Thank you very much for your valuable comments. Lidar measurements in Hong Kong were used to identify the altitude of the volcanic pollutants. Results are shown Supplementary Fig. S5 and in lines 116-122.

> "Lidar measurements in Hong Kong were used to identify the altitude of the volcanic pollutants (Supplementary Fig. S5). As ground-level $PM_{2.5}$ concentrations increased, aerosol concentrations near the surface also rose. Additionally, the majority of volcanic pollutants were transported within the planetary boundary layer, which is approximately 500 meters high. Consequently, these volcanic pollutants significantly affected ground-level air quality in south China."

¬ Figure3: Please show measurements of pollutants from February to May.

Response: Thank you very much for your valuable comments. We extend the duration of the measurements. The new figure shows the measurements of pollutants from March 1 to June 30, 2024. We found that the application of this Chemical-Wind Index (CWI) identified four air pollution events from April to June 2024 (e.g., April 1-2, April 5, April 15-16, and June 21-22) that could be attributed to ongoing emissions from the Taal volcano. During these four pollution episodes, we found unusually high levels of S and P. New results are shown in Figure 3 and in lines 164-171:

> "The results show that the CWI reached a maximum of 73.1 at 6 pm on April 1 (Fig. 3b). Additionally, the application of this index identified three additional air pollution events from April to June 2024 (e.g., April 5, April 15-16, and June 21-22) that could be attributed to ongoing emissions from the Taal volcano. During these four pollution episodes, we found unusually high levels of S (peaking at 18.4 $\mu g/m^3$, 13.6 $\mu g/m^3$, 11.0 $\mu g/m^3$, and 6.8 $\mu g/m^3$), which significantly exceeded its 99th percentile of 4.6 $\mu g/m^3$. Additionally, we found unusually high P levels (peaking at 93.1 ng/m^3, 30.7 ng/m^3, 42.3 ng/m^3, and 38.8 ng/m^3), which also significantly exceeded its 99th percentile of 5.1 ng/m^3."

¬ Providing a more precise explanation of the study's novelty is recommended. What specific contributions does this research make to the existing literature.

Response: We greatly appreciate your comments. We clarify the contributions of our study as follows.

(1) Various studies have used large-scale data to evaluate the impacts of volcanic eruptions by focusing on pollutants that are regularly monitored, such as $PM_{2.5}$ and SO_2. Analysis of the concentration of tracer components in aerosols provides direct evidence of the impacts of volcanic emissions. Detailed measurements of aerosol chemical composition, even at specific locations, can significantly aid in confirming pollution sources during events influenced by volcanoes. In this study, we aim to evaluate the impacts of volcanic emissions on air quality by using a multi-scale data system that combines large-scale data (e.g., satellite monitoring and CTM simulations) with advanced monitoring of aerosol chemical compositions at specific stations in Hong Kong. The real-time multi-scale data were used to identify the potential sources and long-range transport mechanisms responsible for the mysterious air pollution event that occurred in south China on April 1-2, 2024. This multi-scale approach exemplifies comprehensive analyses that accurately attribute air pollution to unusual emission sources, such as volcanoes. Additionally, it enhances emergency preparedness and reduces health risks in regions vulnerable to volcanic events.

We have thoroughly rewritten the Introduction to explain the contribution of our study (lines 64-100).

“Accurately tracking pollution sources for such events requires large-scale data from diverse domains. Akilan et al. (2019, 2023) used GPS data to demonstrate the impacts of volcanic eruptions around the Antarctic continent and in India[9,10]. With improvements in computational capability, state-of-the-art chemical transport models (CTMs) have become advanced tools increasingly used to reveal the transport of air pollutants from volcanoes[11]. Based on a CTM model, Filonchyk et al. (2022) explored the transport of air pollutants from the volcanic eruption in the Canary Islands[12]. Inness et al. (2022) enhanced the capability of another CTM to forecast pollutant transport from the 2019 Raikoke eruption by assimilating SO_2 column and layer height data[13]. Additionally, large-scale satellite measurements have proven useful for revealing emissions and the long-range transport of air pollutants from volcanoes[12,14]. For instance, based on satellite measurements, Milford et al. (2023) evaluated the impact of the 2021 La Palma volcanic eruption on air quality[15].

Most of the aforementioned studies used large-scale data to evaluate the impacts of volcanic eruptions by focusing on pollutants that are regularly monitored, such as $PM_{2.5}$ and SO_2. These large-scale analyses may face two challenges. First, the concentration of $PM_{2.5}$ is influenced by various anthropogenic and natural sources. Therefore, an increase in $PM_{2.5}$ concentration may not necessarily be related to volcanic emissions, complicating the assessment. Second, the concentration of SO_2 may rapidly decrease during long-range transport as it converts to sulfate, which is a component of particulate matter.

Analysis of the concentration of tracer components in aerosols provides direct evidence of the impacts of volcanic emissions[16]. It is widely recognized that volcanoes are significant contributors to the sulfur (S) budget and that volcanic activities lead to increased aerosol S concentrations[19]. Additionally, volcanic ash contains high levels of phosphorus (P), which can promote the growth of vegetation in areas affected by ash deposition[17,18]. During a volcanic event in Hawaii, elevated concentrations of aerosol S and P were detected at the Hawaiian Volcano Observatory[20]. Therefore, detailed measurements of aerosol chemical composition, even at specific locations, can significantly aid in confirming pollution sources during events influenced by volcanoes.

In this study, we aim to evaluate the impacts of volcanic emissions on air quality by using a multi-scale data system that combines large-scale data (e.g., satellite monitoring and

CTM simulations) with advanced monitoring of aerosol chemical compositions at specific stations in Hong Kong. The real-time multi-scale data were used to identify the potential sources and long-range transport mechanisms responsible for the mysterious air pollution event that occurred in south China, particularly in Hong Kong, on April 1-2, 2024. This multi-scale approach exemplifies comprehensive analyses that accurately attribute air pollution to unusual emission sources, such as volcanoes. Additionally, it enhances emergency preparedness and reduces health risks in regions vulnerable to volcanic events."

In the Discussion section, we further discuss impacts of the multi-scale data system (lines 190-197).

> "The analysis in this study highlights the value of integrating multi-scale real-time data capabilities to promptly attribute air pollution events to their sources. The large-scale satellite measurements and model simulations (e.g., CTM and HYSPLIT models) provide a general understanding of transboundary transport. The measurements of aerosol chemical compositions at specific locations offer direct evidence of the impacts on local air quality. The combined analysis of the aerosol chemical composition and large-scale data can help elucidate the potential impacts of unusual emission sources from regions located thousands of kilometers away. "

(2) Another novelty is that we developed a real-time chemical-wind index to identify the occurrence of air pollution associated with volcanic emissions. The development and utilization of the CWI, which integrates local chemical and wind data, provide a robust approach for specific regions to identify the influence of the transported volcanic plumes. The use of local data in the CWI facilitates its application in cities where chemical and meteorological measurements are available. This index is particularly useful for people residing in areas at risk of volcanic activities, helping to protect their public health. We clarify the contributions of the CWI in lines 198-210:

> "Wind conditions in Hong Kong, the Philippines, and even the SCS can affect the transport of air pollutants. In this study, the development of the CWI focuses on its application in a city impacted by volcanic emissions. Therefore, the index relies solely on local data from this city and does not require data from other regions. The development and utilization of the CWI, which integrates local chemical and wind data, provide a robust approach for specific regions to identify the influence of the transported volcanic plumes. The use of

local data in the CWI facilitates its application in cities where chemical and meteorological measurements are available. This index is particularly useful for people residing in areas at risk of volcanic activities, helping to protect their public health. The results of this study support emergency preparedness and help minimize the adverse impacts on the population. Additionally, this prompt analysis demonstrates the scientific process of hypothesis development and testing in near real-time. Such capabilities enhance public understanding of transboundary air pollution through education about the scientific methods used to investigate these events."

¬ The introduction could be improved by incorporating more relevant and recent references.

Response: Thank you very much for your comments. We have thoroughly rewritten the Introduction to incorporate more relevant and recent references. The new paragraphs are shown in lines 64-91:

"Accurately tracking pollution sources for such events requires large-scale data from diverse domains. Akilan et al. (2019, 2023) used GPS data to demonstrate the impacts of volcanic eruptions around the Antarctic continent and in India[9,10]. With improvements in computational capability, state-of-the-art chemical transport models (CTMs) have become advanced tools increasingly used to reveal the transport of air pollutants from volcanoes[11]. Based on a CTM model, Filonchyk et al. (2022) explored the transport of air pollutants from the volcanic eruption in the Canary Islands[12]. Inness et al. (2022) enhanced the capability of another CTM to forecast pollutant transport from the 2019 Raikoke eruption by assimilating SO_2 column and layer height data[13]. Additionally, large-scale satellite measurements have proven useful for revealing emissions and the long-range transport of air pollutants from volcanoes[12,14]. For instance, based on satellite measurements, Milford et al. (2023) evaluated the impact of the 2021 La Palma volcanic eruption on air quality[15].

Most of the aforementioned studies used large-scale data to evaluate the impacts of volcanic eruptions by focusing on pollutants that are regularly monitored, such as $PM_{2.5}$ and SO_2. These large-scale analyses may face two challenges. First, the concentration of $PM_{2.5}$ is influenced by various anthropogenic and natural sources. Therefore, an increase in $PM_{2.5}$ concentration may not necessarily be related to volcanic emissions, complicating the assessment. Second, the concentration of SO_2 may rapidly decrease during long-range transport as it converts to sulfate, which is a component of particulate matter.

Analysis of the concentration of tracer components in aerosols provides direct evidence of the impacts of volcanic emissions[16]. It is widely recognized that volcanoes are significant contributors to the sulfur (S) budget and that volcanic activities lead to increased aerosol S concentrations[19]. Additionally, volcanic ash contains high levels of phosphorus (P), which can promote the growth of vegetation in areas affected by ash deposition[17,18]. During a volcanic event in Hawaii, elevated concentrations of aerosol S and P were detected at the Hawaiian Volcano Observatory[20]. Therefore, detailed measurements of aerosol chemical composition, even at specific locations, can significantly aid in confirming pollution sources during events influenced by volcanoes."

Re: COMMSENV-24-1846-T (*Mysterious air pollution in south China linked to volcanic emissions from the Philippines*)

Dear Editor and Reviewers,

We are grateful to the editor and reviewers for their helpful comments. Modifications have been made in response to these comments. Below is a point-by-point response to each comment. In the responses, text in black represents the reviewers' comments, while text in red represents our responses. A change-tracked version of the manuscript is also uploaded for your reference. We hope that you and the referees find the changes satisfactory, and we look forward to hearing from you soon.

Dr. Changqing LIN, E-mail: cqlin@ust.hk

Division of Environment and Sustainability

The Hong Kong University of Science and Technology

January 15, 2024

Reviewer #1 (Remarks to the Author):

The manuscript is much improved in this version. I have a few minor comments though.

1. Perhaps in the first mention of CTM, indicate that it is based on WRF-CMAQ model so the readers will have an idea that there's chemistry in the model.

Response: We greatly appreciate your comment. We clarify the WRF-CMAQ model used in this study (lines 97-98):

> "The WRF-CMAQ simulations of sulfate (SO_4^{2-}) concentration (Supplementary Fig. S3c) showed a similar track of the volcanic plume."

2. The backward trajectory for 1 April is off the south of Taal, may I suggest that the authors use an ensemble of back trajectories, also with varying heights from near surface to 500m since surface concentrations were also affected, just to reduce the uncertainty of using a single trajectory. This should be easy to redo.

Response: We greatly appreciate your comment. We replot Figure S4 to include the ensemble of back trajectories at 100 m, 200 m, and 300 m above the ground level in Hong Kong on April 1 and 2, 2024, using the HYSPLIT model

3. Can the authors add some more information about the lidar in Fig. S5, can just be included in the figure caption. Information such as general type of lidar, wavelength, resolution, etc.

Response: We greatly appreciate your comment. We add information of the lidar system at the caption of Figure S5:

> "The lidar system, model WINDCUBE 100S, was produced by Leosphere of Vaisala. Measurements with a vertical resolution of 25 m were obtained using a laser beam with a wavelength of 1.543 µm."

4. Line 205: Don't start a paragraph with "however". Rephrase the first sentence.

Response: We greatly appreciate your comment. We revise the sentence accordingly (lines 158-160).

> "We further extend the assessment to the past three years. The application of CWI did not detect any additional significant air pollution cases related to volcanic emissions in Hong Kong during the previous three years."

Also, when submitting a revision, include a clean version and a tracked version.

Reviewer #2 (Remarks to the Author):

The manuscript is acceptable in its current form