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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Nutrients Dissolution Kinetics of Aerosols at Qianliyan Island, the Yellow Sea by a High Time-resolution Nutrient Dissolution Experiment, Potential Linkages with Inorganic Compositions and P solubility controlled factors" *by* Ke Zhang et al.

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Thanks all referees for valuable questions and detail suggestions. Based on all your comments, a detail point-by-point response represented all authors are given as follows.

Response to Anonymous Referee #1

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Discussion paper

1.Comments from Referee: First, the aerosol samples were collected from only one island site and there were no replicates for sampling sites.

Author's response: Just as Jeju island is a representative site in the East Yellow Sea, Qianliyan island is a representative site in the West Yellow Sea, which on the Asia-Pacific atmospheric mass transport path. Besides, island-based investigation has its advantage over the voyage investigation, ensured the continuity of the weekly sampling for all-year observations, which the cruises over the Yellow sea cannot. The unexecuted replicates collection at sampling sites was due to in-sufficient power supply on the island.

Author's changes in manuscript: Add the explanation in 2.1 Site Description, Sample Collection and Sample Selection in manuscript.

2.Comments from Referee: Second, total suspended particulate (TSP) samples were collected by using two different filters in 2011 and 2012 (the poly-carbonate filters in 2011 and Whatman cellulose fiber filters in 2012). I am wondering if these different filters could affect the particle size and composition of the aerosol samples. This should be addressed by an experimental approach.

Author's response: The pore size of the poly-carbonated and the Whatman41 cellulose fiber filter were 0.4μm and 20μm, which may probably cause the missing part of fine particles from atmospheric total suspended particles on Whatman41 filter. However, literatures have reported that Whatman41 filter is suitable for high-volume sampling (Fitzgerald & Detwiler, 1955; Fitzgerald & Detwiler, 1957; Lindeken et al., 1963; Watts et al., 1987; Kitto & Anderson,1988). Particularly for fine particles (submicron aerosols), the collection efficiency was over 75% (Fitzgerald & Detwiler, 1955; Fitzgerald & Detwiler, 1957; Lindeken et al., 1963). Besides, experimental results showed that filter efficiency increase rapidly from 75% to 95% within 25min when aerosol diameter was around 0.4μm and mass concentration was 0.5mgÂům-3 (Lindeken et al., 1963), the filter efficiency was observed to increase rapidly with time. Though the 0.5mgÂům-

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3 is higher than usually encountered in atmospheric monitoring, considered that our sampling time was 20h, coarse particles surely blocked the filter pores, narrowed the pore size during long collection period and our sample TSP mass concentration is higher than usually encountered in atmospheric monitoring, thus our collection efficiency for fine particles was over 95%. Therefore, it has minor filter collection efficiency difference between two filters.

Author's changes in manuscript: Add the explanation in supplementary materials.

3.Comments from Referee: Third, the authors collected 39 aerosol samples but they only measured nutrient dissolution from six aerosol samples. I understand that the authors intended to analyze samples by season and main source. However, it is a pity that the nutrient dissolution experiment has no replicates for each category of samples (spr-SW, spr-NW, sum-NW, aut-NW, win-NW1 and win-NW2). This hinders comparison between sample categories by a statistical analysis.

Author's response: Replicates experiments have been done, which the relative standard deviation (RSD) of concentration of nutrients for the ultra-sound replicates (n=5) were less than 2% and that of high time-resolution dissolution replicates (n=3) (Milli-Q water as leaching solution, 60min) were 1.7%, 1.2%, 2.3% and 3.4% for ammonium, nitrate, phosphate and silicate, respectively. The replicates result for each time point during leaching processing was shown in Figure 1 (in this reply).

Author's changes in manuscript: Add the replicates results in 2.2. Ultra-sound Extraction and Chemical Analysis and 2.3. High Time-resolution Dissolution Experiment in manuscript.

4.Comments from Referee: Moreover, the manuscript also requires extensive editing for English language including grammar and word choice.

Author's response: English writing ability needs to be improved by professional native English speaker.

Author's changes in manuscript: Modify English grammar and word choice throughout manuscript.

5.Comments from Referee: Overall, I don't think this manuscript should be considered for publication at this moment.

Author's response: I hope that my changes will satisfy you.

Author's changes in manuscript: Many changes in manuscript.

6.Comments from Referee: L1 Title: The current title is not appropriate for an ACP submission. It needs to be shortened.

Author's response: I agree that the title need to be shorten.

Author's changes in manuscript: Change the title as Aerosol High Time-resolution Nutrient Dissolution Kinetic, Potential Linkages with Inorganic Compositions and P solubility-controlled factors.

7.Comments from Referee: L26-28: It is likely a speculation that is not well supported by current results.

Author's response: "Compared with the slow dissolution of inorganic P and Si, the rapid dissolution of inorganic N can affect the composition of marine nutrients and marine primary productivity," is not well-expressed. The original intention of this sentence was to express that aerosol, as external source of marine nutrients (N, P and Si), its deposition affect the composition of marine nutrients and marine primary productivity, which is evidence-based speculation. Previous research found that N and P are the main factors driving high level productivity in the Yellow Sea (Gao, 2009; Dou et al., 2011). Aerosol soluble N:P is higher than Redfield ratio, which representing the marine biomasses N: P absorption ratio and the surface water N: P, which representing the marine condition, which correspondingly resulting in N increasement in seawater is significant than P increasement in seawater. Considered that aerosol inorganic N dissolution time (within minutes) and P dissolution time (within hours), aerosol particle

residence time is enough for their soluble phase dissolution. Hence, aerosol deposition affects the composition of marine nutrients and marine primary productivity.

Author's changes in manuscript: Revise this sentence in abstract in manuscript.

8.Comments from Referee: L101-118 (Figure 1): Please give more information on the calculation of backward trajectories, including input data, methodology and validation.

Author's response: The airborne 72h backward trajectory in the intervals of an hour at the height of 1000 m, the lower height of marine atmospheric boundary layer (MABL). All backward trajectories were computed by the Hysplist4 software, which has a wide use in atmospheric transport modeling (Wang et al., 2010; Tošić & Unkašević, 2013; Cohen et al., 2015; Chai et al., 2017). The vertical motion method chose input model data, which were downloaded from NOAA FTP server (arlftp.arlhq.noaa.gov/archives).

Author's changes in manuscript: Provide more information on backward trajectory mapping.

9.Comments from Referee: Figures, 3, 5-8: No replicates and no statistical comparison between aerosol sample categories.

Author's response: Though no replicates for six aerosols, replicate experiments for the ultra-sound and the high time-resolution dissolution experiment have been done. These replicate results basically show better parallelism.

Author's changes in manuscript: No change in manuscript.

Response to Anonymous Referee #2

1.Comments from Referee: The authors discuss the detailed comparisons of a few aerosols from this one site, but most readers will be more interested in the generalities of the results which I think can be summarized as inorganic N dissolves completely and rapidly, while only a percentage of P and Si dissolves and that dissolution takes place over timescales of a few hours.

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Author's response: I agree that more general results should be provided to readers and accepted the referee's proposal.

Author's changes in manuscript: Adjust the descriptive result of the high-time resolution dissolution experiments in Abstract, 3.3. Depictions and Equations for Dissolution Curves and 5. Conclusion in manuscript.

2.Comments from Referee: I would also suggest that the authors need to note a few caveats of this type of experiment. Firstly, wet deposition dominates in most places. The pH of aerosol or rain depositing to seawater will rise to close to 8 almost immediately so prolonged acid exposures can happen in clouds but will then rapidly be reversed. The timescales of dissolution relevant to marine ecosystems are the lifetimes in the surface mixed layer of particles which are days to weeks so the dissolution rates of even the Si and P specie are rapid with respect to that.

Author's response: These caveats should be properly announced in the manuscript.

Author's changes in manuscript: Add the explanation in 2.3. High Time-resolution Dissolution Experiment in manuscript.

3.Comments from Referee: Line 100 Explain why weekly collections only span 20 hours, I assume it is collecting for 1 day each week.

Author's response: The plan sampling time was intended to be one day; however, the actual sampling time was 20 hours per membrane. The aerosol sampler was stopped for one hour every five hours to protect the motor of the sampler and to avoid unstable voltage on the island as well.

Author's changes in manuscript: Add the explanation in 2.1. Site Description, Sample Collection and Sample Selection in the manuscript.

4.Comments from Referee: Line 120 why was 1M HCl used, that is surely very much more acidic. In addition, the P and Si analyses methods are sensitive to the pH of the analyzed solution, did this cause any issues?

Author's response: In ultra-sound extraction, the use of 1M HCl was to obtain the maximum amount of nutrients dissolution compared with aerosol water-soluble nutrient. After acidic extraction and filtration, filtrates were adjusted to neutral used NaOH before the Molybdosilicate Blue methods for P and Si analyses.

Author's changes in manuscript: Add the explanation of the use of 1M HCl in manuscript.

5.Comments from Referee: Line 226 Why do you link the Fe-P pattern to acid processing rather than source, it seems to me it could be either.

Author's response: Fe-P in spr-NW aerosol accounting for 33%, which was higher than Fe-P in its source, Asian sand-dust (0.12%-14%, Yang, Guo & Li, 2012), so except for source, Fe-P pattern also probably linked to the modification in atmospheric transport path (e.g. acid processing).

Author's changes in manuscript: No change in manuscript.

6.Comments from Referee: Line 280-5 and 328-358 These are quite long discussion sections that could be shortened to focus on key and generalizable conclusions rather than specific comparisons of a few aerosols from this one site – consider the wider implications for reader from outside the region.

Author's response: I accept that Line 280-5 and Line 328-358 should be refined.

Author's changes in manuscript: Rewrite 3.3. Depictions and Equations for Dissolution Curves, 3.4. P solubility and 4.1. Relevance of Aerosol Inorganic Components and Dissolution Patterns.

7.Comments from Referee: The issue of comparisons of rates of dissolution for pure minerals, particularly silicates, to the observed rates (around line 380) are an interesting observation that should be retained.

Author's response: I agree to keep the dissolution rate comparison between aerosol

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Discussion paper

and pure mineral.

Author's changes in manuscript: No change in manuscript.

8.Comments from Referee: Line 307 The logic of the discussion around nitrate/ammonium ratios seems to me to be a bit flawed. At the very least it ignores sodium nitrate formed by the sea-salt displacement reaction in the coarse mode aerosol. The nitrate/ammonium ratio in an aerosol depends on emission rates and deposition so I'm not sure this section is particularly useful.

Author's response: The discussion on nitrate/ammonium ratio without Na+ was the most simplified case based on the observation that Na+ was not dominant component in water-soluble components of aerosols along the coastal zone of the Shandong Peninsula, which overlapped the Qianliyan island (Yang & Xiu, 2009). The discussion around nitrate/ammonium ratio have defects in adapting to actual aerosols and its low necessity to the key and general conclusion, therefore this part cannot appear on manusecript.

Author's changes in manuscript: Delete the discussion nitrate/ammonium ratio content.

Response to Williams

1.Comments from Referee: In this study, a series of nutrient dissolution experiments were conducted to determine the soluble fraction of atmospheric nutrients and revealed the short-time dissolution processes, patterns and kinetics of nutrient elements in aerosols. However, I found that the method used to collect aerosol samples had a significant drawback. Total suspended particulate (TSP) samples were collected using poly-carbonate filters in 2011 and Whatman cellulose fiber filters in 2012. Why were different filters used in collecting TSP samples in 2011 and 2012? Was the influence of different filters assessed before the use? This is an important issue because it will largely impact nutrients dissolution kinetics of aerosols and their controlling factors. Supposing both filters are suitable for TSP collection and chemical analysis, but the

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Discussion paper

authors should have used the same kind of filter in the same sampling site for the collection of different season samples. This is essential to maintain the reliability and comparability of the data.

Author's response: The change of filter was a helpless move due to no poly-carbonate filter provider at that sampling period. To keep up the sampling, Whatman41 film had to be used. The pore sizes of the poly-carbonated and the Whatman41 cellulose fiber filter were 0.4μm and 20μm, which might arise fine particles loss on Whatman41 filter. However, former literatures have reported that Whatman41 filter is suitable for high-volume sampling (Fitzgerald & Detwiler, 1955; Fitzgerald & Detwiler, 1957; Lindeken et al., 1963; Watts et al., 1987; Kitto & Anderson,1988). Particularly for fine particles (submicron aerosols), the collection efficiency was over 75% (Fitzgerald & Detwiler, 1955; Fitzgerald & Detwiler, 1957; Lindeken et al., 1963). Besides, experimental results showed that filter efficiency increase rapidly from 75% to 95% within 25min when aerosol diameter was around 0.4μm and mass concentration was 0.5mgÂům-3 (Lindeken et al., 1963), the filter efficiency was observed to increase rapidly with time. Though the 0.5mgÂům-3 is higher than usually encountered in atmospheric monitoring, considered that our sampling time was 20h, coarse particles surely blocked the filter pores, narrowed the pore size during long collection period and our sample TSP mass concentration is higher than usually encountered in atmospheric monitoring, thus our collection efficiency for fine particles was over 95%. Therefore, it has minor filter collection efficiency difference between two filters.

Author's changes in manuscript: Add the information to further explain that filter replacement had less impact.

2.Comments from Referee: Moreover, the sample number 6 in this study is too small to interpret temporal change of aerosol dissolution processes. More than that, I did not find the detailed processes and mechanism on nutrients elements dissolution in aerosols in the present study. The authors should carefully address this issue in more details.

Author's response: Selection of 39 samples was aimed to analyze samples by season and main source. Though sample size was small, they basically captured the seasonal characteristics of the main source direction and satisfied the aim. In this research, the high time-resolution dissolution experiment provides the detailed processes, which cannot get from the most of the total extraction experiments; P form correlated with P dissolution parameters and aerosol P and Si dissolution rates compared with pure minerals provided the potential mechanism of the bulk aerosol, rather than microscopic level mechanism. With the help of sing particle collection, high-resolution electron microscope and other well-developed techniques, more mysteries of microscopic dissolution will be revealed in the near future.

Author's changes in manuscript: Add explanation for choosing six samples.

3.Comments from Referee: Overall, this study is a local investigation in a small island but not focused on studies with general implications for atmospheric science. I do not feel that this manuscript fits the scope of Atmospheric Chemistry and Physics due to its too local interest and its defect in sample collection methods.

Author's response: The Qianliyan island is a representative site in the West Yellow Sea, just as Jeju island is a representative site in the East Yellow Sea, which both on the Asia-Pacific atmospheric mass transport path. It implies that aerosols at this location are both from Asia representing the land and/or the Pacific Ocean representing ocean, which covered large source area. Considered that source effect on aerosol nutrient characteristics (Arnold et al., 1998), the study on aerosol properties at this island has universality in nearby areas. Besides, the issue filter change was explained at first.

Author's changes in manuscript: Add the information on the importance of study area in manuscript.

Response to Zhang

1.Comments from Referee: After reading this manuscript, I ïňĄnd that there are serious

issues in this study. The Qianliyan Island was only one very small sampling site.

Author's response: the Qianliyan island is a representative site in the West Yellow Sea, just as Jeju island is a representative site in the East Yellow Sea, which on the Asia-Pacific atmospheric mass transport path. It implies that aerosols at this location are both from Asia representing the land and/or the Pacific Ocean representing ocean, which covered large source area.

Author's changes in manuscript: Add the information on the importance of study area in manuscript.

2.Comments from Referee: In more marked deïňĄciencies, only 6 aerosol samples were collected during a short period at this small island. Research area is most important for atmospheric science, but I have no idea from this manuscript. Atmospheric Chemistry and Physics (ACP) should publish a large scale study with a wide range of sampling ïňĄelds (e.g., covering the whole Yellow Sea and its coastal areas) and enough samples (e.g., at least 12 months samples which covered a whole year). Therefore, this limited research is not suitable to be published in ACP.

Author's response: Selection from 39 samples was aimed to analyze samples by season and main source, of which seasonal main source can get from daily back trajectories during the collection period. Though sample size was small, they basically represented the seasonal main source/direction characteristics and close backward trajectories for each collection period, which satisfying the aim. Also, the leaching curves of the six aerosols which illustrate that although their absolute concentrations and solubilities are different, their dissolution equilibrium time (different element dissolution time is different, same element dissolution time is similar) and the similar dissolution pattern (dissolution rate goes from fast to slow) has already offered the uniformity rule of aerosol dissolution. From both the ultra-sound extraction and the leaching dissolution experiments, they concluded similar rules for acid stimulation of the same element. In summary, the above experiments meet the origin purpose.

Author's changes in manuscript: Add explanation for choosing six samples and highlight the importance of study area in manuscript.

3.Comments from Referee: These 6 aerosol samples capturing seasonal dominant aerosol sources were selected to carry out the high time-resolution dissolution experiments. It was not clear why only six samples were selected. The 6 aerosol samples are indeed too few to study temporal variation. Moreover, the signiïňĄcance of this research site was not described in detail.

Author's response: The reason for choosing six samples to do the high time-resolution dissolution experiments was to fit the aim of selection that maximized to capture seasonality features of aerosols to the utmost based on the frequency of four directions of seasonal daily back trajectories. The daily -72h back-trajectory of air-masses in spring, summer, autumn, and winter during 2011-2012 at Qianliyan Island were shown in Figure S2 in supplementary material. As for a single sample, the least alternations in source direction were required among around 20 backward trajectories at 1h intervals. In all, 6 samples avoided drastic changes in aerosol source during sampling. The core of this manuscript was not for temporal variation, but the main dissolution characteristics for Asian-West Pacific (terrestrial-ocean) regional aerosol. The main findings of this manuscript are as follows. From both the ultra-sound extraction and the leaching dissolution experiments, they concluded that similar rules for acid stimulation of the same element. It specifically meant that N dissolution was a fast process; while P and Si dissolution was a rather long process and had potential for further release if aerosol met atmospheric acidification and kept the modified soluble phase. From the leaching curves, although aerosol absolute concentrations and solubilities are different, their dissolution equilibrium time (different element dissolution time is different, same element dissolution time is similar) and the similar dissolution pattern (dissolution rate goes from fast to slow), which offered the uniformity rule of aerosol dissolution.

Author's changes in manuscript: Add explanation for choosing six samples in manuscript.

4.Comments from Referee: Sampling information (including sampling location, date, time etc.) of six samples are vital to data analyze and discussion; unfortunately, they were not described.

Author's response: Aerosol sampling information has been partially given in supplementary materials (Table S1) and it will be completed shown in the Table 1.

Author's changes in manuscript: Give the complete information in supplementary materials.

5.Comments from Referee: All data necessary to understand, evaluate, replicate, and build upon the reported research must be made available and accessible whenever possible.

Author's response: All data results from experiments were described, analyzed and concluded. As for replicate, we did replicated experiments for each experiment. The RSD of all nutrient concentrations for the ultra-sound replicates (n=5) were less than 2% and that of high time-resolution dissolution replicates (n=3) (Milli-Q water as leaching solution, 60min) were 1.7%, 1.2%, 2.3% and 3.4% for ammonium, nitrate, phosphate and silicate, respectively. (The replicates result for each time point during leaching processing was shown in Figure 1 (in this reply).) As for data availability, the data of this paper are available upon request (sumeiliu@ouc.edu.cn).

Author's changes in manuscript: Add replicate experiment information in manuscript.

6.Comments from Referee: Critically, I do not see any reference to the air pressure, temperature, wind direction, wind speed, relative humidity, TSP, nutrients concentrations data, not even in the table.

Author's response: Aerosol sampling information (the air pressure, temperature, wind direction, wind speed, relative humidity and TSP) are given in the Table 1 (in this reply) and nutrients concentrations data are given in Figure 3, 4, 5 and 7 in the manuscript.

Author's changes in manuscript: Give the complete information in supplementary ma-

terials.

7.Comments from Referee: Lines 615-623: Three references cite as Chen et al. 2006 were shown. If two or more references from the same year contain the same ïňĄrst six or more authors, use a, b, c, and so on for the in-text citation and in the references list. Please check ACP reference format.

Author's response: It's my mistake and I will correct it immediately.

Author's changes in manuscript: Correct related citations and references.

8.Comments from Referee: It is noted that this manuscript needs careful editing by someone with expertise in technical English editing paying particular attention to English grammar, spelling, and sentence structure so that the goals and results of the study are clear to the reader.

Author's response: Poor English editing will be strengthened afterward.

Author's changes in manuscript: Ask professional native English speaker to correct language mistakes in literature.

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Figure 1. The high-time resolution dissolution curves for replicates (n=3) using accumulated nutrient concentration (error bar: the standard deviation). The leaching solution was Milli-Q water (pH=5.5) and leaching time was 60 min.

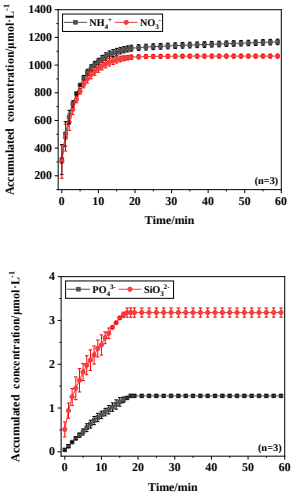

Fig. 1. Figure 1. The high-time resolution dissolution curves for replicates

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Table 1. Sampling date, source, TSP mass concentration ($\mu g \cdot m^{-3}$), mean air pressure (hPa), temperature (°C), wind direction (°) wind speed (m/s) and relative humidity (%) of six aerosols.

Sample	Date	Source	TSP	Air pressure	Temperature	Wind direction	Wind speed	Relative humidity
spr-SW	2011/4/28	SW	35.7	1002.8	11.8	184	5.4	75
spr-NW	2011/3/20	NW	236.4	1011.4	7.6	325	4.5	68
sum-NW	2012/6/12	NW	84.1	994.9	21.8	281	8.0	84
atu-NW	2012/10/13	NW	80.4	1011.2	20.5	309	9.4	60
win-NW1	2011/2/12	NW	56.5	1019.8	-2.2	338	4.2	70
win-NW2	2011/1/28	NW	129.3	1025.0	-1.6	314	5.6	52

Fig. 2. Table 1. Sampling information