Correspondence to Review 2

Thank you very much for your thorough and constructive comments on our manuscript amt-2020-10, entitled "A semi-continuous study on the toxicity of atmospheric particles using a versatile aerosol concentration enrichment system (VACES): development and field characterization". We made all corrections and revised the manuscript according to your comments. The response is given to each comment. In the revised manuscript, changes including some technical corrections are colored in blue.

General comments: The manuscript represents the results obtained by using a versatile aerosol concentration enrichment system (VACES), which was extended in order to estimate the toxicity of ambient air. The differences between concentrated and non-concentrated particulate matter were manifested in the measured aerosol properties; while offline and online sampling equipment has been applied to investigate in detail the acquired toxicity data. The observed results were further enhanced by studying the correlations between the measured toxicity and all obtained physical/chemical aerosol parameters, in order to highlight the role of aerosol enrichment through the VACES technique. Overall, the results are interesting and well investigated but the manuscript is difficult to follow. An important vantage of this study is that it includes a variety of techniques, providing several aerosol properties that may capture the effect of VACES on sampled particulate matter. Finally, it is essential that this study attempts to highlight the role of aerosol chemical components on the measured toxicity levels. Nevertheless, the manuscript needs some major revision regarding the selected toxicity assay, the fact that the utilized techniques are not clearly described and, in several places the justifications or discussions should be presented in a more comprehensive way.

Response: Thank you for your recognition of our results and overall research work. In response to your professional opinions, we have made major revisions to the deficiencies in the original manuscript, including the detailed explanation of toxicity assay, more specific description of experimental techniques, rephrasing the justifications and discussions so that it is clear.

Major issues:

Comment 1: Section 1: Please justify the application of photobacteria in atmospheric particle toxicity and its association with health effects.

Response 1: Comment 1 is an important question, we reply as follows,

First is the application of photobacteria in atmospheric particle toxicity (exactly ecotoxicity, we changed toxicity to ecotoxicity in response to Reviewer 1). In fact, photobacteria have long been used in the

study of particle ecotoxicity and even related to specific chemical components in ambient air. Take the researches in recent years as an example, Turóczi et al. (2012) used *Vibrio fishcer* to study the ecotoxicity of PM_{10}. This study directly evaluated the overall ecotoxicity of particles from different sources and seasons. Tositti et al. (2018) developed an ecotoxicity detection method using *Vibrio fishcer*, and found that ecotoxicity was closely related to the compositions of PM_{10}. Wang et al. (2016) demonstrated that the $PM_{2.5}$ components analyzed by *Photobacterium Phosphoreum T3* bioassay was ecologically toxic. Eck-Varanka et al. (2019) analyzed the ecotoxicity of size-fractionated particles using *Vibrio fischeri*. Such literature proved the feasibility of the photobacteria-based method in assessing the ecological toxicity of atmospheric particulate matter. The relevant description of the ecotoxicity assay of PMs in previous studies were summarized and added in manuscript (Introduction section).

Second, it remains a scientific issue in vitro experiments that there is a lack of direct data support of the relationship between toxicity (e.g., cytotoxicity and ecotoxicity) and adverse PM health effects. Even for the exposure experiments (e.g., fish and mammalian), to our knowledge, no study exposes animals and human simultaneously to PMs due to ethics. Therefore, we removed all contexts on health effects and emphasized on the measurements and data to avoid from over-extrapolating the impacts and implications of the results to human health (as Reviewer 1 mentioned). Even so, the method of using photobacteira bacteria to study ecotoxicity or even cytotoxicity is feasible because: 1) the method of measuring ecotoxicity using photobacteria has long been routinely applied for water and soil research. This method has been standardized by the International Standards Organization (ISO 21338:2010: Water quality - Kinetic determination of the inhibitory effects of sediment, other solids and colored samples on the light emission of *Vibrio Fischeri*/ kinetic luminescent bacteria test; 2) many applications in previous studies on particle ecotoxixity as above mentioned; 3) there are also several studies reported strong correlations between the Microtox (Photobacterium phosphoreum) EC_{50} and rat/mouse LD_{50} values (e.g., Fort, 1992; Kaiser et al., 1994). The information was also summarized in manuscript (Introduction section)

References

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Tositti, L., Brattich, E., Parmeggiani, S., Bolelli, L., Ferri, E., and Girotti, S.: Airborne particulate matter biotoxicity estimated by chemometric analysis on bacterial luminescence data, Sci. Total Environ., 640, 1512-1520, 2018.

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Wang, W., Shi, C., Yan, Y., Yang, Y., and Zhou, B.: Eco-toxicological bioassay of atmospheric fine particulate matter (PM2.5) with Photobacterium Phosphoreum T3, Ecotox. Environ. Safe., 133, 226-234, 2016.

Comment 2: Section 2.2: It is difficult to follow the description and the setup of the instrumentation. Please rephrase to clarify the definition of the optimal technical parameters, the calculation of measured theoretical concentrations, the utility of ambient aerosol removal through filter, the description of performance testing, the essential technical difference between offline and online collection, the justification of selected sampling parameters (both offline and online). Please rephrase to describe the obtained samples since there are collections mentioned as continuous, based on 1min, on 8hrs, on 1hr and on 30min.

Response 2: As pointed out in Comment 2, we rephrased Methodology, and all above mentioned points in bold were explained in details and in a better arrangement. Moreover, in order to look more intuitive and clearer and easier to explain, we modified the set-up structure in Figure 1 and added necessary legends and a full explanation in the caption of the figure.

The revised Methodology in blue:

2 Methodology

2.1 Design of VACES

VACES used a saturation and condensation system to rapidly grow particles into super-micron droplets, which were then concentrated by a virtual impactor. Detailed description of the design of VACES is available in previous studies (e.g. Kim et al., 2001a, b). Briefly, when the airflow was sucked into a water tank filled with deionized water (defined as a saturator) with a U-shaped heating tube inside, the particles became supersaturated. A tube was fixed above the outlet of the saturator, and a copper tube coil was tightly wound on the outside to provide fast condensation conditions. A chiller (Bilon, China) filled with ethanol (80 %, Hushi, China) cooled through the coil. The condensed aerosols were drawn up to a virtual impactor, where particle concentration in sizes was concentrated to a desired level by changing the ratio of the major-to-minor air flow controlled by a mass flow controller (MFC, D08-4F, Sevenstar, China).

2.2 Sampling

Sampling was conducted for several experiments, including laboratory performance test, field performance test, discontinuous sample collection and continuous sample collection. **The performance test** in this study used the **enrichment factor (EF)** defined as the ratio of **concentrated (VACES) to non-concentrated (ambient)** particle concentration, and **the enrichment efficiency (EE) defined as the ratio of the concentrated concentration to ten times the non-concentrated concentration** as a standard. The closer the EF and EE are to 10 and 100%, respectively, the better the enrichment effect of VACES. The instrument operating **parameters** (major air flow, minor air flow, condensation temperature and saturation temperature) were defined as **the optimal parameters** when the best enrichment effect was obtained. In the **laboratory performance test**, an atomizer (Model 9302, TSI, USA) was used to atomize polystyrene latex (PSL, Thermo Fisher Scientific, USA) to produce 200 nm, 300 nm, 500 nm and 700 nm particles respectively (Figure 1). On the one hand, after drying the generated PSL particles (Nafion tube, MD-700, Perma Pure, USA), set the corresponding voltage through a differential mobility analyzer (DMA, Model 3081, TSI, USA) for screening, and then entered the condensation particle counter (CPC, model 3775, TSI, USA) at a flow rate of 0.3 L min^{-1} for counting. On the other hand, PSL particles were introduced into VACES to obtain ten times the concentrations, and then the concentrations were calculated by the system of

DMA and CPC after drying. Use four set data (number and mass) of PSL particles with and without enrichment to draw the EF calibration line. In comparison, **field performance test** was similar to laboratory test. The only **difference** was the replacement of PSL particles with ambient particles (<1.0 µm). In addition, during the field performance test, an **aerosol filter** (ETA Filters, USA) was installed at the inlet of the saturator to remove ambient particles to study the formation of

115 particles in the VACES (Fig. 1; the dash line marked the filter location). Then, if no or few particles were observed in CPC, the impact of particle formation in VACES could be excluded or ignored.

In the performance test, we determined the optimal parameters (as defined above) of VACES. Then, we successively carried out **discontinuous and continuous VACES particle collection** on the sixth floor of the Environmental Science and Engineering Department of Fudan University in Shanghai. We opened the inlet to the ambient air, in which particles were

120 sucked into the saturator at a major flow rate and increased in concentration at a minor flow rate (Kim et al., 2001a). VACES particles were collected in 5 ml of deionized water through a biosampler (SKC, USA) for 30 minutes and 1 hour. In order to study the physio-chemical and ecotoxicity differences between VACES particles and environmental particles, we switched the inlet of the biosampler to ambient air after VACES particles were collected, that is, 30 minutes (1 hour) VACES samples, then 30 minutes (1 hour) environmental samples. From October 23 to December 11, 2019, we obtained a total of 10 sets of

125 30-minute samples and 10 sets of 1-hour samples. Therefore, due to time discontinuity, sampling was defined as **discontinuous collection**. In contrast, in the continuous sample collection process, we added a $PM_{2.5}$ sampler (PM-100, Wuhan Tianhong, China) to the inlet of VACES. To achieve **continuous VACES particle collection**, we added a peristaltic pump (BT100-4, HUXI, China) forward of the biosampler to pump in deionized water, and connected the outlet of biosampler to another peristaltic pump to evacuate the sample into an automatic fraction collector (BS-40A, HUXI, China).

130 Pumping in and out were performed at a scheduled time (59-minute sleep mode and 1-minute work mode) and volume (5 mL). However, **the continuous collection of ambient particles** was performed by a $PM_{2.5}$ cyclone (Met one Instruments, USA) at a flow rate of 5 L min^{-1} and on a 47 mm Teflon filter (Whatman, USA). The filter sampling time is 8 hours to meet the detection limit of water-soluble ions. In the end, we collected 88 VACES samples and 11 simultaneous ambient samples from December 18th to 31st, 2019. Note that the filter samples were extracted in 10 ml deionized water via 20-minute

135 sonication and < 34 °C (< 34 °C within filter bottle and 45 °C out of filter bottle) heating condition.

2.3 Measurements

All samples were filtered using 0.22 µm pore size filters (Collins, China) and 10ml sterile syringes (KDL, China). Then, ecotoxicity assay and water-soluble ion measurement were conducted immediately. Regarding to the ecotoxicity assay, Jing et al. (2019) provided detailed information. In brief, 100 µL of the prepared bacterial suspension was pipetted into cuvettes

140 to measure the luminous intensity as the baseline. After, the initial luminous intensity was recorded after adding 100 µL of sample. In 15 minutes, the luminous intensity was recorded again. After subtracting blank intensity tested using NaCl solution (3 %), the light inhibition rate of VACES and ambient particles was calculated, respectively, according to the international standard (ISO 11348-1: 2007) procedures (Water quality, 2007). All samples were tested in triplicate and averaged in present study. To ensure the enrichment effect of VACES system, we also detected water-soluble ions of both

145 ambient and VACES samples collected during continuous sampling period using an ion chromatography (940 Professional IC Vario, Metrohm, Swiss) integrated with an autosampler (863 Compact Autosampler, Metrohm, Swiss). Moreover, the atmospheric $PM_{2.5}$ concentration was monitored in a nearby state-controlled site (Liangcheng, Hongkou, Shanghai, China).

The revised Figure 1 below:
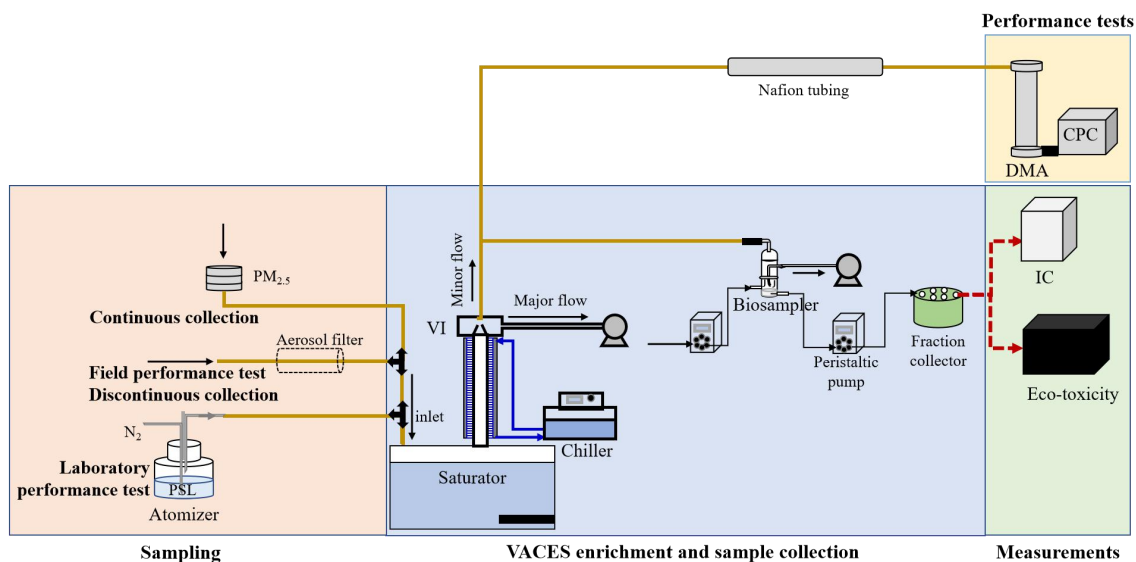
Figure 1 Set-up for performance test and field sample collection. The experiments referring to VACES including Laboratory and field performance tests, discontinuous and continuous sample collection and their measurements. In laboratory performance test, air flow passed through atomizer, VACES (saturator-condensation tube-virtual impactor), nafion tubing, DMA, and CPC successively; For two field performance tests, in the first one air flow passed through VACES, nafion tubing, DMA, and CPC successively, and in the second one air flow passed through aerosol filter, VACES, nafion tubing, DMA, and CPC successively; During discontinuous sample collection, particles followed the flow line of VACES-to-biosampler; For continuous sample collection, particles were collected from $PM_{2.5}$ sampler, VACES, biosampler, to fraction collector. Both types of collected samples were used for online ionic measurement by an ion chromatography and online eco-toxic assay by a photobacterium acute toxicity analyzer (an integrated instrument with automatic operation controlling by a programming).

Comment 3: Section 2.3: As already mentioned in Section 1, the selected toxicity assay is mentioned in Jang et al., (2019) as a biotoxic assay based on a luminescent bacterium. Please justify the selection of this particular assay and clarify how this could be linked to health effects. Please mention whether this assay has been part of epidemiological studies or has been compared / combined with any cellular or acellular aerosol toxicity assay linked to health effects.

Response 3: For the first question, the reason why we selected the biotoxicity test based on luminescent bacteria is that the detection method is rapid and the luminescent bacteria are sensitive to changes in the concentration of toxic components in particles. The luminescent bacteria assay is an acute toxicity detection method. Its maximum exposure time is 24 hours, and its adverse effect (bacterial death) should occur within 14 days. The ecotoxicity determination in this study could be completed within 15 minutes, while the culture time of the bacteria was only 5 minutes. In addition, Figure 5 in the original manuscript showed that changes in ecotoxicity are very sensitive to changes in $PM_{2.5}$. In order to achieve online ecotoxicity measurement, we decided to use this method initially.

Regarding the second question, this is a toxicology study, which is not directly related to health effects in this study or previous studies revised in response 1. Therefore, in order to focus on the results and avoid exaggerating the significance of this study, we have deleted all discussions about health effects.

Regarding the third question, there is still a lack of research reports based on the direct relationship between the ecotoxicity of bacteria and the health effects of atmospheric particles. Because low concentrations of atmospheric particulate matter require a long exposure time to have health effects, but bacteria are very sensitive and their exposure time is less than 24 hours (that is, the big difference in exposure time will make it difficult to conduct correlation studies between the two). However, several studies have reported a close correlation between Microtox (phosphobacteria) EC50 and rat/mouse LD50 values (for example, Fort, 1992; Kaiser et al., 1994), as described in response 1.

All the replies mentioned above are already in the "Introduction" section or have been added. The blue font below shows better arrangements and revisions to the introduction:

1 Introduction

Currently, most toxicological studies focus on discovering the relationship between particulate matter and the morbidity or mortality of organisms (e.g. Vincent et al., 2001; Cox et al., 2016; Miri et al., 2018), or exploring toxic mechanisms by exposure experiments (e.g. Magnani et al., 2016; Huang et al., 2017; Rychlik et al., 2019). However, the measurement ecotoxicity data are rarely available because of technical limitations. For instance, it requires a long detection time due to the animal and plant reproduction or cell cultivation (National Research Council, 2006), but the concentration and chemical composition of particulate matter in the atmosphere continue to change over time, especially during severe pollution (Shang et al., 2018a, b). Thereby, a short analyzing time is quite important.

To solve this problem, photobacteria (e.g. Photobacterium phosphoreum) are utilized in the ecotoxicity study of atmospheric particles, because the detection was rapid (e.g. within 15 minutes; Hoover et al., 2005) and the cultivation period is only 5 minutes (Jing et al., 2019). The method of measuring ecotoxicity using photobacteria bioluminescence inhibition bioassay has long been routinely applied and standardized for water and soil research (ISO 21338:2010: Water quality – Kinetic determination of the inhibitory effects of sediment, other solids and coloured samples on the light emission of Vibrio fischeri /kinetic luminescent bacteria test/). It had been reported that the photobacterium phosphoreum EC_{50} (median effective concentration) significantly correlated to rat and mouse LD_{50} (the lethal dose for 50 percent of the animals tested) values, indicating the reliability of photobacteria-based ecotoxicity assay (Fort, 1992; Kaiser et al., 1994). Recently, photobacteria have also been often used to assess the ecotoxicity of particulate matter and chemical components in atmosphere. For instance, Turóczi et al. (2012) used *Vibrio fishcer* to study the ecotoxicity of PM_{10}. This study directly evaluated the overall ecotoxicity of particles from different sources and seasons. Tositti et al. (2018) developed an ecotoxicity detection method using *Vibrio fishcer*, and found that ecotoxicity was closely related to the compositions of PM_{10}. Wang et al. (2016) demonstrated that the $PM_{2.5}$ components analyzed by *Photobacterium Phosphoreum T3* bioassay was ecologically toxic. Eck-Varanka et al. (2019) analyze the ecotoxicity of size-fractionated particles using *Vibrio fischeri*. Such literature proved the feasibility of the photobacteria-based method in assessing the ecological toxicity of atmospheric particulate matter. However, the detection limit of ecotoxicity using photobacteria is high. For example, in Jing's research, samples with a light inhibitory rate of less than 20 % were considered to be non-toxic due to the impact of normal bacteria fluctuations. Whereas, the concentration of atmospheric aerosols is usually far lower than that required for eco-toxic assay in case of short sampling time (e.g. one hour), which means a longer sampling time is required. Nevertheless, long-time sampling may lead to a large loss of volatile substances or chemical reactions in the particles, subsequently resulting in large errors in ecotoxicity analysis (Weiden et al., 2009).

215 In this respect, aerosol enrichment techniques have been developed and applied to increase aerosol concentrations to meet ecotoxicity detection limits……

225 **Response 4:** According to EPA standards, $PM_{2.5}$ does not exceed 35 $\mu g/m^3$ in a 24-hour period. Therefore, our original classification standard is that the days less than or equal to 35 $\mu g/m^3$ are clean days, and the days greater than 35 $\mu g/m^3$ are polluted days. However, due to the limitation of the amount of data, original manuscript did not perform a divisional chemical or ecotoxicity analysis on clean and polluted days. Therefore, we deleted the information of pollution classification and only
230 retained the $PM_{2.5}$ concentration range to emphasize that even at low PM concentrations, this integrated detection method could also detect its ecotoxicity.

The "non-ambient samples" was changed to "VACES samples" which was defined in revised Methodology section (with ten times the concentration enrichment in VACES).

We changed the expression from "normal fluctuation range of the luminescence bacterium affecting the
235 sample uncertainty" to "Note that since bacteria is sensitive to environmental condition, it is difficult to determine whether the sample is non-toxic or toxic as inhibition rate is lower than the baseline" for better understanding.

When the concentration was low, the change in the light inhibition rate of ambient particles did not match the concentration of environmental $PM_{2.5}$. However, under high concentrations of $PM_{2.5}$, they
240 matched better. During continuous sampling period, $PM_{2.5}$ concentrations varied from 14 to 107 $\mu g\ m^{-3}$. The light inhibitory of both ambient and VACES particles exhibited similar trends with the change of $PM_{2.5}$ concentration (a strong positive correlation, r2> 0.7) (Fig. 5). Note that as removing the data points of low $PM_{2.5}$ concentration, the correlation coefficient increased. Both experiments indicated that as the concentration of $PM_{2.5}$ decreased, the impact of concentration on ecotoxicity might be weakened,
245 and the ecotoxicity may be caused mainly by the toxic chemical components in the particles (Akhtar et al., 2014). In this regard, the effects of key toxic components on ecotoxicity changes need to be further studied using VACES.

The change of EF was roughly reversed to the trend of the light inhibition rate of the ambient and VACES particles (Fig. 4). The main reason was that the increase of the light inhibition rate of VACES
250 particles was lower than that of the ambient at high $PM_{2.5}$ concentrations.

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Comparing the EF changes of chemical components, it was found that when the ratio of light inhibition rate of VACES to ambient particles decreased, the EF corresponding to nitrate also decreased (Fig. 6). It showed that under high PM$_{2.5}$ concentrations, the EE of VACES for high-concentration nitrate was reduced, which was probably attributed to the loss of nitrate in VACES at a higher saturation temperature (about 7 °C higher than the ambient). According to calculations, among the VACES particle concentration loss (average of 20.1 % during entire experiment period), nitrate accounted for 18.0 %. Therefore, under the premise of providing sufficient water vapor, reducing the saturation temperature or reducing the deviation from the ambient temperature were an important way to improve the enrichment effect of VACES.

The arranged and detailed (in blue) Discussion and Conclusion section were showed as follows:

3.2 Ecotoxicity variation of VACES particles

The study evaluated the ecotoxicity by the light inhibition rate of photobacteria, the higher the value, the higher the ecotoxicity. The light inhibition rate was calculated by one hundred multiplying the ratio of the difference in fluorescence intensity between treated and untreated medium to the untreated medium, where untreated medium meant only bacteria in medium without particle samples, treated medium corresponded to a sample adding in bacteria medium. Discontinuous sampling was operated under PM$_{2.5}$ concentrations ranging from 21 to 187 μg m^{-3}. During the sampling period, the photobacteria light inhibition rate of almost all ambient samples was lower than the baseline (20 %). On the contrary, the rate of all VACES samples was higher than 20 % (Fig. 4a and 4b). Note that since bacteria is sensitive to environmental condition, it is difficult to determine whether the sample is non-toxic or toxic as inhibition rate is lower than the baseline. It implied that the increase in ecotoxicity caused by increase of particulate concentration could avoid the inaccurate assessment of particulate ecotoxicity in the range below the baseline. In addition, when the concentration was low, the change in the light inhibition rate of ambient particles did not match the concentration of environmental PM$_{2.5}$. However, under high concentrations of PM$_{2.5}$, they matched better. During continuous sampling period, PM$_{2.5}$ concentrations varied from 14 to 107 μg m^{-3}. The light inhibitory of both ambient and VACES particles exhibited similar trends with the change of PM$_{2.5}$ concentration (a strong positive correlation, r^2> 0.7) (Fig. 5). Note that as removing the data points of low PM$_{2.5}$ concentration, the correlation coefficient increased. Both experiments indicated that as the concentration of PM$_{2.5}$ decreased, the impact of concentration on ecotoxicity might be weakened, and the ecotoxicity may be caused mainly by the toxic chemical components in the particles (Akhtar et al., 2014). In this regard, the effects of key toxic components on ecotoxicity changes need to be further studied using VACES.

The change of EF was roughly reversed to the trend of the light inhibition rate of the ambient and VACES particles (Fig. 4). The main reason was that the increase of the light inhibition rate of VACES particles was lower than that of the ambient at high PM$_{2.5}$ concentrations. Comparing the EF changes of chemical components, it was found that when the ratio of light inhibition rate of VACES to ambient particles decreased, the EF corresponding to nitrate also decreased (Fig. 6). It showed that under high PM$_{2.5}$ concentrations, the EE of VACES for high-concentration nitrate was reduced, which was probably attributed to the loss of nitrate in VACES at a higher saturation temperature (about 7 degrees higher than the ambient). According to calculations, among the VACES particle concentration loss (average of 20.1 % during entire experiment period), nitrate accounted for 18.0 %. Therefore, under the premise of providing sufficient water vapor, reducing the saturation temperature or reducing the deviation from the ambient temperature were an important way to improve the

enrichment effect of VACES.

4 Conclusions and implications

To achieve detection limits for atmospheric particulate ecotoxicity, a versatile aerosol concentration enrichment system (VACES) was extended to be integrated with ecotoxicity measurement. The VACES was developed to increase particle concentrations by about 7–10 times under the conditions of chiller temperature (-19 ± 1 °C), saturator temperature (45 ± 2 °C, supersaturation temperature was less than 31 °C), major air flow (50 ± 1 L min^{-1}), and minor-to-major flow ratio (1/10). We conducted discontinuous and continuous sample collection to analyze the ecotoxicity of VACES and ambient particles in half-hour and one-hour time resolution, respectively. It was found that the ecotoxicity of almost all ambient samples below the detection baseline as ambient PM$_{2.5}$ concentration varied from 14 μg m^{-3} to 187 μg m^{-3}. After enrichment, however, the ecotoxicity was significantly detected for almost all samples, proving the feasibility of the integrated system on rapid ecotoxicity assay. In addition, by comparing the change of the ambient PM$_{2.5}$ concentration with the light inhibition rate of ambient and VACES particles, it was found that as the concentration of PM$_{2.5}$ decreased, the correlation between the PM$_{2.5}$ concentration and the light inhibition rate was significantly weakened. It meant that at low concentrations of particulate matter, the impact of concentration on ecotoxicity was greatly reduced, and the interference with ecotoxicity might be the change of toxic components. Moreover, during the high PM$_{2.5}$ concentration period, the ecotoxicity of VACES particles and the EF were significantly weakened or reduced, due to the loss of nitrate in relatively high-temperature VACES system, which provided a guidance for improving the enrichment effect of VACES.

In addition, apart from the upper revision of Figure 1, we also added some legend explanation in figure captions like:

Figure 4 Comparison of light inhibition rate and ratio of ambient and VACES particles with ambient PM$_{2.5}$ concentration based on (a) hourly and (b) 30 min discontinuous sample collection during 23rd October–11st December, 2019 in Shanghai, China. Baseline reflected the accuracy of photobacteria based ecotoxicity assay method and below the baseline, the accuracy is low.

Figure 5 Comparison of light inhibition rate between ambient and VACES particles based on continuous sampling of VACES and ambient. VACES samples were collected hourly and ambient filter samples were collected every eight hours. The PM$_{2.5}$ concentration data was collected hourly from a nearby monitoring center (online data).

Figure 6 Enrichment factors of chemical compositions and light inhibitory of PM$_{2.5}$ during continuous sampling period. The EF was calculated by the ratio of chemical concentrations of VACES to ambient particles. The component concentration of VACES particles was one hour per sample, and the concentration of ambient particles was 8 h per sample. For the ratio, we averaged the concentrations of VACES samples every 8 h to correspond to that of ambient samples.