ABSTRACT  Nitrotriphenylenes (NTPs), which include the highly mutagenic isomer 2-nitrotriphenylene (2-NTP), have been detected in airborne particles. From a public health point of view, it is necessary to study the environmental occurrence of NTPs in detail. In this study, concentrations of five nitrated polycyclic aromatic hydrocarbons (nitro-PAHs) including NTPs in airborne particles and of nitrogen oxides (NOx; NO+NO2) and carbon monoxide (CO), at a location in South Osaka, Japan, were measured at 3 h intervals. It was found that the diurnal variations in the concentrations of 1-nitropyrene (1-NP), NOx, and CO were similar, being high early in the morning and late in the evening. This finding indicates that the occurrence of 1-NP is affected significantly by primary emissions, particularly by automotive emissions. The concentration change in 1-nitropyrene was similar to that of 2-nitropyrene produced by an atmospheric OH radical-initiated reaction. On the contrary, the variations in the concentrations of 2-nitrofluoranthene (2-NF) and 2-NTP were significantly different from those of the other nitro-PAHs, i.e., their concentrations increased during the nighttime, suggesting that neither 2-NF nor 2-NTP was emitted from the primary sources, but were formed via the NO3 radical-initiated nitration of the parent fluoranthene and triphenylene (TP) in the atmosphere. Based on the ambient concentration of 2-NTP and the reported rate constant for the reaction of TP with NO3 radicals, the yield of 2-NTP from the gas-phase NO3 radical-initiated reaction of TP was estimated to be 23%.

KEY WORDS  PAH, Nitroarene, Triphenylene, Secondary formation, Airborne particles

1. INTRODUCTION

Most nitrated polycyclic aromatic hydrocarbons (nitro-PAHs), a class of polycyclic aromatic compounds (PACs), exhibit direct-acting mutagenicity or carcinogenicity (Durant et al., 1996). Some of them have been detected in particulate matters in the ambient air (Bamford and Baker, 2003) or combustion exhaust (Schuetzle et al., 1982). For example, 1-nitropyrene (1-NP) has been detected in both diesel exhaust particles (Schuetzle, 1983) and airborne particles (Feilberg et al., 2001). Although 1-NP can be formed from gas-particle phase heterogeneous reactions on a specific substrate such as mineral dust aerosols (Kameda et al., 2016) or under the high NO2 condition (Nguyen et al., 2009; Miet et al., 2004;
Kameda et al. (2005), which include the strong mutagenic isomer 2-nitrotriphenylene (2-NTP) (Zimmermann et al., 2012; Kameda et al., 2006), which include the strong mutagenic isomer 2-nitrotriphenylene (2-NTP) (Ishii et al., 2001, 2000). The rate constants for the gas-phase reactions of TP with OH or NO3 radicals have been determined by a relative-rate technique in a CCl4 liquid-phase system (Kameda et al., 2013). Because strongly mutagenic 2-NTP has been found in the air at concentrations comparable to those of 1-NP and 2-NF, which are the most abundant airborne nitro-PAHs (Zimmermann et al., 2012; Kameda et al., 2006; Kawanaka et al., 2005; Kameda et al., 2004; Ishii et al., 2001; Ishii et al., 2000), its contribution to the mutagenicity of airborne particles is expected to be significant. However, it is difficult to understand the factors influencing the formation and behaviour of atmospheric NTPs, because they are produced from not only the gas-phase radical-initiated reactions but also gas-soluble-phase heterogeneous reactions of TP with NO2/NO3/N2O5 (Yang et al., 2013; Ishii et al., 2000) and combustion of organic matters such as fossil fuels (Zimmermann et al., 2012; Kameda et al., 2006). From a public hygienic point of view, the acquisition of more detailed data about the environmental occurrence of NTPs is of great importance.

In the present study, we sampled airborne particles at a point in South Osaka, Japan at 3-hr intervals, and determined the particle-associated NTPs, other typical nitro-PAHs, and gaseous atmospheric pollutants simultaneously in order to understand the factors controlling the occurrence and concentrations of atmospheric NTPs.

2. MATERIALS AND METHODS

2.1 Sampling of Airborne Particles
Airborne particulates were collected on quartz fiber filters (Advantec MFS, QR100) using high-volume air samplers (Kimoto Electrics, Model 120) on the rooftop of a three-story building (approximately 10 m above the ground level) at Osaka Prefecture University in Sakai, Osaka, Japan (34°55’N, 135°1’S1’E). The sampling site was located in a slightly polluted residential area in South Osaka, which can be regarded as a typical suburban area in Japan. Vehicular traffic on moderately busy roads, Route 310 and Hanwa-Highway, 200 m and 2 km away from the sampling site, respectively, was the only substantial source of air pollutants; no other significant potential stationary source of air pollution was present near the site. Air samples were collected at intervals of 3 h over two 27-hr periods: (I) September 4 and 5, 2001 and (II) November 28 and 29, 2001. The average air collection rate of the air samplers was 1.5 m³ min⁻¹ and no cut-off stage was employed. The filter samples were stored at 253 K until they were subjected to the analysis.

2.2 Chemical Analysis of Nitro-PAHs
The filter samples were cut into fine pieces, and all the fine pieces were put into 200 mL of benzene/ethanol (3/1, v/v). Soluble organic fractions (SOF) was extracted by sonication for 15 min twice, then the extract solution was filtered to remove solid substances with cellulose acetate filter (Advantec MFS, No.2). The filtrate was washed with 100 mL of 5% sodium hydroxide, 100 mL of 20% (v/v) sulfuric acid and 100 mL of water. After evaporating the organic layer to ca. 5 mL and filtered with a 0.22 μm membrane filter, the solvent was gently removed with a nitrogen stream. A 0.5 mL sample solution was obtained by adding methanol to the residue. An aliquot of the sample solution was injected into the HPLC system. The HPLC system with column-switching and chemiluminescence detection was employed for nitro-PAHs quantification as reported previously (Kameda et al., 2006; Kameda et al., 2004). Although nitro-PAHs are not fluorescent,
they are metal-catalytically reduced to strongly fluorescent amino-PAHs in the HPLC system, enabling a sensitive and selective detection of nitro-PAHs by the chemiluminescence detector. For the calibration curves of the standard nitro-PAHs, the chemiluminescence intensities were proportional to the concentrations of the compounds up to 2000 fmol per injection, and the calibration curves showed good linearity \((r > 0.999)\). The quantification limits of the HPLC system employed for 1-NP, 2-NP, 2-NF, 1-nitrotriphenylene (1-NTP), and 2-NTP were 2, 20, 10, 75, and 60 fmol, respectively \((S/N = 3)\). Chemical structures of the target nitro-PAHs in this study are summarized in Table 1 with the information of their atmospheric sources.

### 2.3 Gas Analysis

During the sampling period, typical gaseous pollutants, namely, \(\text{NO}_x\) (\(\text{NO} + \text{NO}_2\)) and CO were monitored with a chemiluminescence \(\text{NO}_x\) analyzer (Thermo Electron, MODEL 42s) and an NDIR CO analyzer (Thermo Electron, MODEL 48), respectively.

### 2.4 Chemicals

1-NP was obtained from Sigma-Aldrich Co; 1-NTP and 2-NTP were obtained from Hayashi Pure Chemical Ind. Ltd. and ChemBridge Co., respectively; and 2-NF and 2-NP were purchased from Chiron AS. All the solvents and other chemicals used were HPLC or analytical grades from Wako Pure Chemical Ind., Ltd.

### 3. RESULTS AND DISCUSSION

Table 2 shows the concentrations of selected inorganic gases and nitro-PAHs in 3-hr averaged samples of the airborne particles obtained in this study. The diurnal change of 1-NP in airborne particles collected between November 28 and 29 (abbreviated as Nov 28–29) is shown in Fig. 1a with the concentrations of \(\text{NO}_x\) (= NO + \(\text{NO}_2\)) and CO, which were measured during the same period of time. The concentrations of \(\text{NO}_x\) and CO were in the ranges from 13 to 53 ppbv and from 0.6 to 1.3 ppmv, respectively. These concentrations can be regarded as normal for residential areas with slight pollution in Japan (Morikawa et al., 1997) and the observed concentrations of 1-NP associated with airborne particles \((29–96 \text{ fmol m}^{-3})\) were comparable to values reported by other researchers in Japan (Kamiya et al., 2017; Kojima et al., 2010a; Ishii et al., 2001), Hong Kong (Ma et al., 2016), New Zealand (Kalisa et al., 2019), and Czech Republic (Nežíková et al., 2020). The diurnal variations in concentrations of particle-associated 2-NF, 2-NP, 1-NTP, and 2-NTP on Nov 28–29 are shown in Fig. 1b. The concentration of 2-NF ranged from 33 fmol m\(^{-3}\) for the sample collected between 12:00 and 15:00 (abbreviated as 12:00) to 412 fmol m\(^{-3}\) for the sample collected at 0:00. The concentration of the 2-NF obtained in this study is substantially the same as the previously reported levels in Japan (Kamiya et al., 2017; Kojima et al., 2010a; Kojima et al., 2010b), France (Albinet et al., 2007), and Italy (Di Filippo et al., 2010). The concentration of 2-NTP

### Table 1. Structures of nitro-PAHs observed in this study and their atmospheric sources.

| Nitro-PAHs | Atmospheric Sources |
|-----------|---------------------|
| 1-nitropyrene (1-NP) | Primary emission from combustion sources, gas-solid-phase heterogeneous reaction (minor contribution) |
| 2-nitropyrene (2-NP) | Gas-phase OH radical-initiated reaction |
| 2-nitrofluoranthene (2-NF) | Gas-phase OH radical-initiated reaction, gas-phase \(\text{NO}_3\) radical-initiated reaction |
| 1-nitrotriphenylene (1-NTP) | Primary emission from combustion sources, gas-solid-phase heterogeneous reaction, gas-phase OH radical-initiated reaction |
| 2-nitrotriphenylene (2-NTP) | Primary emission from combustion sources, gas-solid-phase heterogeneous reaction, gas-phase \(\text{NO}_3\) radical-initiated reaction |
was also the lowest (10 fmol m\(^{-3}\)) at 12:00 and reached the maximum (43 fmol m\(^{-3}\)) at 0:00. On the contrary, the highest concentrations of 1-NTP and 2-NP were observed at 18:00. The concentrations of 1-NTP and 2-NP were in the ranges from 8 to 72 fmol m\(^{-3}\) and from 21 to 86 fmol m\(^{-3}\), respectively. The observed concentration of 2-NP in airborne particles was slightly higher than that measured previously in Japan (Kamiya et al., 2017; Murahashi et al., 1999), France (Ringuet et al., 2012), and Italy (Di Filippo et al., 2010). While the concentrations of 1-NTP and 2-NTP obtained in this study were lower than previously reported levels in Tokyo, Japan (Ishii et al., 2001; Ishii et al., 2000), these values were slightly higher than those reported in Riverside, CA and Mexico City, Mexico (Zimmermann et al., 2012). The diurnal variations in the concentrations of NO\(_x\), CO, and 1-NP, which are primarily emitted from the combustion processes, were similar, i.e., their concentrations increased early in the morning and late in the evening. However, the concentrations of 2-NF and 2-NTP were low during the daytime and high at night. The opposite diurnal variability of 2-NF and 2-NTP compared with the diurnal variability of the primary emitted chemicals clearly indicates that 2-NF and 2-NTP did not originate from the primary combustion sources, but were formed by the atmospheric nitration of the parent FLRA and TP, respectively. 2-NF was found to be produced from the gas-phase reactions initiated by NO\(_3\) or OH radicals in the presence of NO\(_x\) (Atkinson and Arey, 1994; Atkinson et al., 1990; Arey et al., 1986). At night, the atmospheric concentration of OH radicals, which are produced from photochemi-

Table 2. Concentrations of selected inorganic gases and nitro-PAHs in 3-hr averaged samples of the airborne particles collected in Osaka, Japan on September 4–5, 2001 and November 28–29, 2001.

| Date               | Time\(^{a}\) (JST) | Nitro-PAHs\(^{b}\) | Inorganic gases |
|-------------------|---------------------|-------------------|----------------|
|                   | 1-NP  | 2-NP  | 2-NF  | 1-NTP | 2-NTP | CO\(^{c}\) | NO\(_x\)\(^{d}\) |
| September 4–5, 2001 |       |       |       |       |       |       |       |
| 12:00             | 17    | 4     | 26    | N.A.  | 9     | 0.3    | 37    |
| 15:00             | 22    | 18    | 11    | N.A.  | 13    | 0.3    | 54    |
| 18:00             | 43    | 35    | 108   | N.A.  | 16    | 0.6    | 47    |
| 21:00             | 34    | 52    | 187   | N.A.  | 16    | 0.7    | 44    |
| 0:00              | 33    | 27    | 369   | N.A.  | 31    | 0.6    | 48    |
| 3:00              | 28    | 17    | 293   | N.A.  | 31    | 0.5    | 44    |
| 6:00              | 54    | N.A.  | 553   | N.A.  | 36    | 0.7    | 66    |
| 9:00              | 17    | 3     | 176   | N.A.  | 22    | 0.4    | 32    |
| 12:00             | 40    | 1     | 63    | N.A.  | 17    | 0.3    | 28    |
| Mean              | 32    | 20    | 199   | N.A.  | 21    | 0.5    | 45    |
| November 28–29, 2001 |       |       |       |       |       |       |       |
| 12:00             | 87    | N.A.  | 33    | N.A.  | 10    | 0.6    | 13    |
| 15:00             | 29    | 21    | 64    | 22    | 16    | 0.7    | 22    |
| 18:00             | 82    | 86    | 168   | 72    | 23    | 1.1    | 46    |
| 21:00             | 96    | 61    | 272   | 32    | 31    | 1.3    | 53    |
| 0:00              | 41    | 49    | 412   | 12    | 43    | 1.0    | 35    |
| 3:00              | 37    | 34    | 313   | 8     | 26    | 0.8    | 20    |
| 6:00              | 59    | 76    | 239   | 12    | 27    | 1.1    | 35    |
| 9:00              | 47    | 26    | 81    | 8     | 20    | 0.8    | 17    |
| 12:00             | 43    | 52    | 201   | 33    | 20    | 0.9    | 43    |
| Mean              | 58    | 51    | 198   | 25    | 24    | 0.9    | 32    |

Abbreviations of compounds: see text and Table 1.
NA: not available
\(^{a}\)Start of sample collection
\(^{b}\)Given in unit of fmol m\(^{-3}\)
\(^{c}\)Given in unit of ppmv
\(^{d}\)Given in unit of ppbv
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cal processes, is reduced almost to zero. On the contrary, NO$_3$ radicals generally increase after sunset, and play a significant role in the atmospheric nighttime reactions. The nighttime increase in the 2-NF concentration observed in this study suggests the significance of the gas-phase NO$_3$ radical-initiated nitration of FLRA. The increase in 2-NF during the nighttime could also be partly attributed to the transportation of the air mass containing 2-NF secondarily produced from the OH radical-initiated reaction of FLRA during the daytime. However, the concentration of 2-NP, which is probably not formed by an NO$_3$ radical-initiated reaction but by an OH radical-initiated reaction (Atkinson and Arey, 1994) in the gas-phase, did not increase during the nighttime. This supports the idea that a significant part of 2-NF was produced by the reaction of FLRA with NO$_3$ radicals in the gas phase, at least over the duration of the sampling in this study. 2-NTP was previously reported to be formed by a gas-

Fig. 1. Diurnal variations in the concentrations of 1-NP (solid triangle), NO$_x$ (cross), and CO (open diamond) (a), and 1-NTP (solid diamond), 2-NTP (open square), 2-NF (open circle), and 2-NP (plus) (b) in south Osaka, Japan on November 28–29, 2001.

solid phase heterogeneous reaction (Yang et al., 2013; Ishii et al., 2000) or a gas-phase homogeneous reaction (Kameda et al., 2006) of the parent TP. Although both reactions, namely, the reaction of the TP deposited on a solid-surface with NO$_3$/NO$_2$/N$_2$O$_5$ and that of the gas-phase TP with OH radicals in the presence of NO$_2$, yield 2-NTP, these processes can also yield abundant amounts of 1-NTP (Yang et al., 2013; Zimmermann et al., 2012; Kameda et al., 2006; Ishii et al., 2000). However, the gas-phase reaction of TP with NO$_3$/NO$_2$/N$_2$O$_5$ produces almost only 2-NTP (Kameda et al., 2006). Therefore, the observed diurnal variation in 2-NTP in this study, which was similar to that of 2-NF and different from that of 1-NTP, strongly suggests that a significant part of 2-NTP in airborne particles collected during the sampling period was formed from the gas-phase NO$_3$ radical-initiated reaction of TP in the atmosphere. We have previously demonstrated that the correlation between the atmospheric concentrations of 2-NTP and 2-NF ($r = 0.70$, significance level $p < 0.001$) was significantly better than the correlation between 2-NTP and 2-NP ($r = 0.38$, $p < 0.001$) (Kameda et al., 2005). These correlation coefficients were based on more than 90 data sets. This result also supports the view that NO$_3$ chemistry can yield atmospheric 2-NTP.

Changes in the concentrations of 1-NTP were accompanied by similar changes in the concentration of 2-NP (Fig. 1b). When the concentration of 2-NP increased, the increase was probably because of only the OH chemistry. Therefore, atmospheric 1-NTP was assumed to be formed mainly by an OH radical-initiated reaction; however, some part of it may have come from direct emis-

Fig. 2. Diurnal variations in the concentrations of 1-NP (solid triangle), 2-NP (plus), 2-NTP (open square), and 2-NF (open circle) in south Osaka, Japan on September 4–5, 2001.
The diurnal changes in the nitro-PAH concentrations in airborne particles collected on September 4–5 (abbreviated as Sept 4–5) (Fig. 2) and Nov 28–29 (Fig. 1) were similar, i.e., the concentrations of 1-NP and 2-NP were low in both daytime and nighttime, whereas those of 2-NF and 2-NTP were elevated in the nighttime. The 1-NTP concentration could not be determined during the Sept 4–5 period owing to the lack of the sample amount.

When all the data obtained on Sept 4–5 and Nov 28–29 were combined, the 2-NF concentration increased linearly with an increase in the 2-NTP concentration (r = 0.91, p < 0.001), although the x-intercept value was significant (Fig. 3a). When the daytime (6:00–15:00) and nighttime (18:00–3:00) data were plotted separately, the x-intercept was significant in the daytime (Fig. 3b), but almost zero in night (Fig. 3c). This result implies that the potential sources of 2-NTP, for example, direct emissions, existed during the daytime and that the nighttime formation pathways of 2-NTP and 2-NF were the same or almost similar. The mean value of the observed concentration ratio of [2-NF]_obs/[2-NTP]_obs during the nighttime determined by a least-square analysis was 10.1. The rate of the atmospheric concentration change of 2-NF and 2-NTP can be given by the following equations based on their rate of formation by the NO3 radical-initiated reaction:

\[
\frac{d[2-NF]}{dt} = y_{2NF} k_{FLRA-NO3}[NO3][FLRA] - k_{loss-2NF}[2-NF] \quad (1)
\]

\[
\frac{d[2-NTP]}{dt} = y_{2NTP} k_{TP-NO3}[NO3][TP] - k_{loss-2NTP}[2-NTP] \quad (2)
\]

where \( y_{2NF} \) and \( y_{2NTP} \) are, respectively, the yields of 2-NF and 2-NTP from the gas-phase reactions of FLRA and TP with NO3 radicals; \( k_{FLRA-NO3} \) and \( k_{TP-NO3} \) are the rate constants of the reactions; \( k_{loss-2NF} \) and \( k_{loss-2NTP} \) are, respectively, the rate constants for the atmospheric loss processes of 2-NF and 2-NTP such as photolysis and oxidation; and \([FLRA]\) and \([TP]\) are the gas-phase concentrations of FLRA and TP, respectively. Under steady state conditions, both these rates are equal to zero. Therefore, the ratio of the loss rate constant \( k_{loss-2NTP}/k_{loss-2NF} \) can be expressed as follows:

\[
k_{loss-2NTP}/k_{loss-2NF} = \frac{y_{2NTP} k_{TP-NO3}[TP][2-NF]_{ss}}{y_{2NF} k_{FLRA-NO3}[FLRA][2-NTP]_{ss}} \quad (3)
\]

where \([2-NF]_{ss}\) and \([2-NTP]_{ss}\) are the steady state concentrations of 2-NF and 2-NTP, respectively. The concentrations of particle-associated FLRA and TP at the sampling site were reported to be comparable (Kameda et al., 2004). Significantly less literature is currently available on the gas-particle distribution of TP. Lammel et al. (2010a, 2010b) reported the particle-phase mass fractions of TP (\( \theta_{TP} \)) and FLRA (\( \theta_{FLRA} \)) to be 0.52–0.63 and 0.07–0.12, respectively, at ambient temperatures of 289.4–309.0 K. Therefore, assuming that the particle-phase concentrations of TP and FLRA were the same,
the gas-phase concentration ratio \([\text{TP}]/[\text{FLRA}]\) was estimated to be \(-0.08\) using the mean \(\theta_{\text{TP}}\) and \(\theta_{\text{FLRA}}\) values. The reaction rate constant \(k_{\text{FLRA-NO}_3}\) was determined to be \(5.1 \times 10^{-28}[\text{NO}_2] \text{ cm}^3\text{molecule}^{-1}\text{sec}^{-1}\) and \(y_{2\text{NF}}\) was determined to be \(24\%\) (Atkinson and Arey, 1994). Although the reaction rate constant \(k_{\text{TP-NO}_3}\) was determined to be \(0.66 \times 10^{-28}[\text{NO}_3] \text{ cm}^3\text{molecule}^{-1}\text{sec}^{-1}\) (Kameda et al., 2013), the yield of 2-NTP from the gas-phase \(\text{NO}_3\) radical-initiated reaction has not been obtained. The relative loss rate \(k_{\text{loss-2NTP}}/k_{\text{loss-2NF}}\) was estimated to be \(-0.1\) based on the ambient concentrations of \(\text{TP}, \text{FLRA, 2-NTP, and 2-NF}\), the rate constant for the reaction of \(\text{TP}\) with \(\text{OH}\) radicals, and the rate constant for the reaction of \(\text{FLRA}\) with \(\text{OH}\) radicals assuming the steady state condition as well as Eq (3) (Kameda et al., 2013). Therefore, using the reported reaction rate constants and assuming that \([2\text{-NF}]_i/[2\text{-NTP}]_i = 10.1\), \(k_{\text{loss-2NTP}}/k_{\text{loss-2NF}} = 0.1\), and \([\text{TP}]/[\text{FLRA}] = 0.08\), Eq (3) gives the yield \(y_{\text{2NTP}}\) of 23%. The yield of 2-NTP from the reaction of \(\text{TP}\) with \(\text{NO}_3/\text{NO}_2/\text{N}_2\text{O}_5\) in \(\text{CCl}_4\) was determined to be \(35\%\) (Kameda et al., 2006). The nitro-isomer profiles of the reactions of several PAHs with \(\text{NO}_3/\text{NO}_2/\text{N}_2\text{O}_5\) in \(\text{CCl}_4\) were similar to those in the gas-phase \(\text{NO}_3\) radical-initiated reactions (Phousongphouang and Arey, 2003; Zielinska et al., 1986). In those studies, the yield was higher than 20% for several types of nitro-PAH isomers in the gas-phase \(\text{NO}_3\) radical-initiated nitrations, which is in agreement with our estimation of the yield of 2-NTP. The abundance of 2-NTP in the atmosphere, in spite of the low reactivity of the parent \(\text{TP}\) (Kameda et al., 2013; Nielsen, 1984), may be attributed not only to the high stability of 2-NTP (i.e. the small \(k_{\text{loss-2NTP}}\) value) but also to the high yield of 2-NTP from the gas-phase \(\text{NO}_3\) radical-initiated reaction of \(\text{TP}\).

4. CONCLUSIONS

In this study, diurnal concentrations of NTPs in airborne particles at a location in South Osaka, Japan were monitored at 3-hr intervals over two 27-hr periods. The concentration change in 1-NTP was very similar to that of 2-NP produced by an atmospheric OH radical-initiated reaction. On the contrary, the variations in the concentrations of 2-NF and 2-NTP were significantly different from those of other nitro-PAHs, i.e., they increased during the nighttime, suggesting that 2-NF and 2-NTP were formed via the \(\text{NO}_3\) radical-initiated nitrations of the parent FLRA and TP in the atmosphere. Based on the ambient concentration of 2-NTP and the reported rate constant for the reaction of \(\text{TP}\) with \(\text{NO}_3\) radicals, the yield of 2-NTP from the gas-phase \(\text{NO}_3\) radical-initiated reaction of \(\text{TP}\) was newly determined to be 23%. This high yield may be one of the reasons for the high concentration of 2-NTP in the atmosphere. Although the observation period in this study was not recent, the results obtained in this study on the secondary formation of NTPs via the radical-initiated reactions can be considered to be general and independent of the observation year. Despite the strong mutagenicity of 2-NTP, very few studies have been performed to measure its concentration in the atmosphere. Although, in this study, we successfully estimated one of the factors that led to the high concentration of 2-NTP, further elucidation of the atmospheric behavior including the occurrence and loss processes of NTPs is desired.

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