The Atmospheric Concentration of Common Greenhouse Gases in the Pulpwood Plantation in Riau Province, Indonesia

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Abstract. Pulp and paper wood plantations are one of the substantial forestry estates in Indonesia that are usually integrated with a pulp and paper mill industry. The atmospheric condition of glasshouse gases that keep increasing primarily due to anthropogenic activities represent critical issues on human health and affect forest productivity. Therefore this study measured the atmospheric concentration of common glasshouse gases in the pulpwood plantation integrated with a pulp mill in Riau Province. Overly high concentration of some glasshouse gases in the atmosphere may cause issues and at an extreme level, may lead to the occurrence of acid rain that could affect forest health and productivity. This study employed the Ogawa Passive Sampler Method to determine the air concentration of common glasshouse gases, i.e., [SO₂],[NOₓ],[NO₂], and [O₃] in different rotations and stand ages of Acacia crassicarpa and Eucalyptus pellita stands. The results showed that species, rotation age, and stand age affect the concentration of gaseous N in the plantations. The concentrations of [NOₓ] and [NO₂] gases were higher in the old rotation than in the young rotation of A. crassicarpa stands. Those concentrations were also relatively higher in peat soil sites than in non-peat soil sites.

1. Introduction
The air concentration of common greenhouse gases, i.e., CO₂, O₃, NOₓ, SO₂, CH₄, and others, has been increasing sharply due to many factors, e.g., anthropogenic activities, which develops threat to human health as well as affect forest productivity. Ozone in the atmospheric layer that poses dangerous effects is called tropospheric O₃. The concentration of this tropospheric O₃ is found to increase rapidly in Northeast Asia [1,2]. The World Meteorological Organization (WMO) recognized the importance of those atmospheric gases monitoring, thus initiated the Global Atmosphere Watch (GAW) to monitor the atmospheric condition of the world.

In order to succeed, forest plantations should consider the effect of the increasing atmospheric pollutants, e.g., ozone, SO₂, and NOₓ, on the prospective plantation species such as eucalypts and acacias. The plantations of eucalypt and acacia have been well-established to supply materials for pulpwood, sawlog, furniture, fuelwood, and other wood-based industries. The genera’s fast-growing traits and wide range of adaptability make them potential plants to mitigate climate change as well as to provide wood supplies for various wood-related industries. In order to determine the atmospheric condition at the field, the atmospheric [O₃], [SO₂], [NO₂], and [NOₓ] in the eucalypt and acacia...
plantations in Riau Province, Indonesia, were measured and discussed in this study. The presence of greenhouse gases in large amounts could harm humans, vegetation, and the environment. For example, nitrogen; it is common knowledge that deposited N in the soil is favorable for plants because it is an essential nutrient required by plants. The exceeded amount of NO\(_2\) and NO\(_x\) than the required level in the atmosphere may disturb the natural N cycle and it may, in turn, harm the plants and cause a loss in biodiversity. NO\(_x\) is also free radical gas that could directly affect plants physiologically by biological reactions within leaf cells [3]. The chemical reaction in the atmosphere converts NO to NO\(_2\), which further can result in O\(_3\) [3]. SO\(_2\) is also known to be harmful to plant, specifically when it exposes in an exceeding amount and a long duration of time. Exposure to high doses of SO\(_2\) can lead to necrosis on leaves that would further impair growth and could finally lead to plant death. The concentration of SO\(_2\) in the atmosphere has been found to increase in many countries lately [3]. This condition should raise a concern about its effects on plants and the environment. Combination of SO\(_2\) and NO\(_2\) (or NO\(_x\)) was evidently reduced chlorophyll content and growth of wheat and mustard plants [4].

Ozone (O\(_3\)) in the troposphere layer, in particular, is well known as a highly oxidative gas and could be detrimental to both humans and plants. O\(_3\) in exceeding amount could inhibit plant growth by a reduction in photosynthetic rate due to damage on leaves caused by the oxidative gas [5–7]. The recently increased concentrations of atmospheric pollutant gas (i.e., SO\(_2\), NO\(_x\), and O\(_3\)) in many developing countries, especially Asia, can cause severe damage to vegetation, and such damage is likely to worsen in the future as pollutant emissions continuously increase [8]. Thus, it is vital to recognize the effect of those gases on eucalypts and acacias. Knowledge on the current performance of eucalypts and acacias in the plantation or the air quality of the forest could give information on whether the forest already faced environmental threats. That information might give further perspectives on the possible future condition of the eucalypts and acacias in the plantation. Therefore the atmospheric N gaseous and ozone concentration in Indonesia (tropical region), i.e., Riau Province, were measured. For comparison, the gases concentration was also measured in Taiwan (sub-tropical region), i.e., Taichung City. Thus, the observation was aimed to get a brief insight on field gaseous atmospheric conditions in the acacia and eucalypt plantations, whether it already possessed environmental threats to the plants or not. Full-scale investigation on environmental conditions is undoubtedly essential, but the full-scale examination was only conducted on controlled growth chambers and was discussed in separate papers. However, this result may provide preliminary insight for a further full-scale investigation of environmental conditions in the field.

In the locations of measurements, i.e., Indonesia and Taiwan, industrialization is increasing at a rapid pace. In Indonesia, the deforestation rate is also high. Furthermore, Taiwan is surrounded by heavily industrialized countries, e.g., China, Hong Kong, Japan; thus, we assumed that the concentration of air pollutant gases might not be far different between the two countries.

2. Materials and Methods

2.1. Measurement sites
The measurement was conducted in Perawang city (1° 4’N; 102° 52’ E), Riau Province in Indonesia; and in Taichung city (24° 09’ N; 120° 40’ E) in Taiwan. For Indonesia, two city sites (downtown and nursery) and eight forest sites were measured, while one city site and one forest site (two replication) were measured in Taiwan.

The sites in Indonesia represent two species which were *E. pellita* and *A. crassicarpa*. While *E. pellita* was grown in two types of soil, non-peat soil (histosols) and peat soil (contain organic matters), *A. crassicarpa* was only grown in peat soil. Measurements were conducted in old age stand (>3 years old) and young stand (< 6 months old). Especially for *A. crassicarpa*, the measurement between old and young rotations was also compared. Here, rotation is a term for the harvest cycle in the plantation. One rotation consists of 5 years, which means the stand is harvested five years after planting in the site. Sites in Taiwan were planted with *Acacia confusa* and are not for commercial purposes.
2.2. The assembly of passive sampler and gas concentration analysis

The ambient air gases concentration was measured by a Passive Sampler Method (Ogawa & Co. USA, Inc) [9]. The gases measured were air ambient of [O₃], [SO₂], [NO₂], and [NOₓ]. The average air temperatures were 29.5°C and 18.8°C in Indonesia sites and Taiwan sites, respectively.

![Figure 1](image)

**Figure 1.** The Ogawa passive sampler components: body (6) 20 mm outer diameter, 15 mm inner diameter, 30 mm length; diffuser end caps (1); stainless screens (2); collection filters 14.5 mm diameter (3); Teflon ring (4) and Teflon disc (5) (source: Ogawa & Co. USA, Inc).

The Ogawa passive sampler components were assembled in accordance with orders in figure 1. Each passive sampler for [O₃], [NOₓ], [NO₂], and [SO₂] were equipped with collection filters coated with specific chemicals to trap the target gases. The filter in the O₃ sampler was coated with nitrite, while Triethanolamine coated the filter for NO₂ and SO₂ sampler, and Triethanolamine plus PTIO (2-phenyl-4,4,5,5-tetramethylimidazoline-3-oxide-1-oxy) for NOₓ. The assembled samplers were kept in tight-sealed plastic bottles to avoid contamination from air or other contaminants until measurement. To trap the target gases, the passive samplers were set at a particular height in the relatively open area (canopy free) and were exposed to the air for at least one week. After exposure time was completed, the sampler was collected and were kept again in tight-sealed bottles to avoid contamination until extraction and analysis with ion chromatography.

In the laboratory, the samplers were disassembled and the filters were subsequently extracted with suitable reagents. Milli-q water was used to extract O₃. For analysis of NO, NOₓ, and NO₂, those reagents were prepared: 80 g sulfanilamide in a mixture of 200 ml concentrated phosphoric acid and 700 ml water; 0.56 g N-(1-Naphthyl)-ethylenediamine dihydrochloride in 100 ml water (NEDA solution); 1.5 g dried-sodium nitrite (dry over 4 hours, at 105-110°C) in 1 liter water; and nitrite standard solutions from concentrations of 0-1.0 µg nitrite/ml water. For analysis of [SO₂], 1.75% solution of hydrogen peroxide was used to extract it from the filter.

The concentration of the gases was determined with ion chromatography in the laboratory of Environmental Conservation Division, Institute of Environmental Science, Hokkaido Research Organization, Sapporo, Hokkaido, Japan.

The calculation for the gases concentration in ambient air was in accordance with the following equations:

\[
\begin{align*}
[O_3] \ (ng \ g^{-1}) &= \alpha_{O3} \times W_{O3} / t, \\
[SO_2] \ (ng \ g^{-1}) &= \alpha_{SO2} \times W_{SO2} / t, \\
[NO_x] \ (ng \ g^{-1}) &= W_{NOx} / t, \\
[NO_2] \ (ng \ g^{-1}) &= \alpha_{NO2} \times W_{NO2} / t, \\
[NO] \ (ng \ g^{-1}) &= \alpha_{NO} \times (W_{NOx} - W_{NO2}) / t,
\end{align*}
\]

where \( W_{O3} \), \( W_{SO2} \), \( W_{NOx} \), and \( W_{NO2} \) are O₃, SO₂, NOₓ and NO₂ quantity (respectively) from the collection filter, determined by ion chromatography; \( \alpha_{O3}, \alpha_{SO2}, \alpha_{NOx}, \alpha_{NO2}, \) and \( \alpha_{NO} \) are conversion coefficient for (ng g⁻¹) concentration; \( t \) is exposure time. The conversion coefficients are as follows:

\[
\alpha_{O3} = 46.2 \times 10^2 \times (293 / (273 + T))^{0.3} / (9.94 \times \ln (t) - 6.53),
\]
\[ \alpha_{SO_2} = 39.4 \times \frac{293}{(273 + T)}^{1.83}, \]
\[ \alpha_{NO_2} = \frac{10000}{(0.677 \times [P] \times [RH]) + (2.009 \times [T]) + 89.8}, \]
\[ \alpha_{NO} = \frac{10000}{(-0.78 \times [P] \times [RH]) + 220}, \]
\[ [P] = \left( \frac{2P_n}{P_T + P_n} \right)^{2/3}, \]

\( T \) is the ambient temperature (°C); \( t \) is the exposure time (min); \([RH]\) is the relative humidity (%); \( P_n \) is 17.535 (water vapor pressure in mm Hg at 20°C); \( P_T \) is the vapor pressure of water at the ambient temperature \( T \).

3. Results and Discussion

3.1. Air quality in the eucalypt and acacia plantation

The [NO\(_x\)] and [NO] in \( A. crassicarpa \) stands were higher in the old rotation than in the young rotation (figure 2). Within the same rotation, the young stand had a relatively higher concentration of those gases than the old stand. The [NO\(_x\)], [O\(_3\)], and [SO\(_2\)] were relatively the same between the old and young rotations (figure 2). The [O\(_3\)] in the young stand was relatively higher than that in the old stand. The [SO\(_2\)] was low in all of the observed sites. The concentrations of [NO\(_x\)] and [NO] in the old rotation of \( A. crassicarpa \) were evidently higher than those in the young rotation and \( E. pellita \) sites (figure 2).

The [NO\(_x\)] and [NO] were higher in the peat soil stand of \( E. pellita \) than those in the non-peat soil stand (figure 2). Unlike \( A. crassicarpa \) in the peat soil, the old age stand of \( E. pellita \) in the peat spoil had higher [NO\(_x\)] and [NO] than the young stand, while the results were opposite in the non-peat soil stands. The [NO\(_x\)], [O\(_3\)], and [SO\(_2\)] were relatively the same between peat soil and non-peat soil, a trend that was relatively similar to those in the \( A. crassicarpa \) stands. However, unlike the [O\(_3\)] in the peat soil of \( A. crassicarpa \), the concentration in the peat soil stand of \( E. pellita \) was higher in the old age stand than in the young stand (figure 2).

![Figure 2](image-url)

**Figure 2.** The concentration of air pollutant gases collected with passive samplers in \( Acacia crassicarpa \) stands (A) and in \( Eucalyptus pellita \) (B), between stand consist of old age (> 3 years old) and young age trees (< 6 months old) in Indonesia. A shows the comparison between old rotation (rotation 4 = the land had been used for 20 years) and young rotation (< rotation 1, which means the land had been used less than 5 years) of \( A. crassicarpa \) stands. B shows the comparison between \( E. pellita \) stands in the non-peat and peat soil.
A. crassicarpa is a species in the genus Acacia that can survive in the wetland and strong-acidic soil [10]. Therefore, this species has been planted in peatland by most plantation forestry companies for pulpwood supplies in Indonesia. The concentrations of N-derived gases were higher in A. crassicarpa stands than those in E. pellita stands (figure 2 and 3). There are two possible explanations for that result, organic matters in the peatland and the N-fixing capacity of A. crassicarpa. The higher concentration of N-derived gases (especially NO and N\textsubscript{2}) in the atmosphere was probably due to the contribution of nitrification (in the form of N\textsubscript{2}O and NO) and denitrification (in the form of N\textsubscript{2}O, NO, and N\textsubscript{2}). Moreover, plants in genus Acacia are acknowledged for the symbiosis with the nitrogen-fixing microbes and mycorrhizae; thus, the soil in which those plants grow may contain a high concentration of N [11–13]. Therefore, A. crassicarpa stands showed a higher concentration of [NO\textsubscript{x}], [NO\textsubscript{2}], and [NO] than E. pellita stands.

The old rotation of A. crassicarpa had higher [NO\textsubscript{x}] and [NO] than the young rotation (figure 2). These acacias were grown in the peat soil, which means the plants grew above organic materials. The old rotation had a higher concentration of the gases, presumably due to the higher amount of organic matter accumulation in the soil during the duration of the rotation. The organic matter was accumulated from harvesting wastes (e.g., branches, barks, leaves, and roots), added to the already high organic matter of the peat soil. The older rotation had a higher number of harvests and more considerable waste biomass on the site. The older the rotations, the higher the volume of organic matter would be accumulated in the soil. The same reason might also explain the higher concentration of those gases in the peat soil stand of E. pellita than in the non-peat soil stand. Furthermore, [14] found that when N is abundantly available or is not a limiting nutrient, the changes in greenhouse gas emissions concerning the future higher temperature and vegetation composition are depended on the N-availability.

Irrespective of rotation, the young stand of A. crassicarpa had higher [NO\textsubscript{x}] and [NO] than the old stand. The open-air area is wider in the young stands (due to the smaller canopy) than in the old stands and that means a broader area is exposed to sunlight, leading to higher air temperature than in the old stands. The air temperature in the area might be related to the age and structure of the stands. There is a possibility that high temperature might increase the amount of N-derived gases emitted from peat soil [15].

However, the trend was different in the peat soil stand of E. pellita, in which the concentration of the respective gases was higher in the old stand than those in the young stand. The rotation of the E. pellita stands was a rotation 4. Likely, the age of rotation had more significant impact on the gases emission than the age of the stand.
Figure 3. The concentration of air pollutant gases taken in the commercial plantation in Indonesia. A shows the average gases concentration in the city, nursery, and in the stands of E. pellita and A. crassicarpa; meanwhile, B shows the average gases concentration in the city, nursery, and stands at the mineral and peat soil.

Ozone concentration in the atmosphere is related to the concentration of N-derived gases. Thus if the concentration of N-derived gases was relatively higher in one location than the other, then there is a big possibility for \([\text{O}_3]\) in the former-mentioned location to be higher than in the latter. The \([\text{O}_3]\) in A. crassicarpa stands was higher in the young stand than that in the old one; in contrast, it was lower in the young peat soil stand of E. pellita. These results suggest that in peat soil stands of E. pellita, the open-air area did not increase the gases emission. The rotation of E. pellita was rotation 4; therefore, the age of rotation may be more influential on the gases emission than the age of the stands.

As expected, a higher concentration of N-derived gases (\(\text{NO}_x\), \(\text{NO}_2\), and \(\text{NO}\)) was observed in the city sites than those in the nursery and forest sites, but \([\text{O}_3]\) was somewhat similar among all sites (figure 3). The \([\text{NO}_x]\) and \([\text{NO}]\) were relatively higher in the peat soil sites than those in the non-peat soil sites (figure 3). Anthropogenic activities are busier in the city, whereas there are more significant numbers of vehicles, factories, and all activities that contribute to the higher concentration of those gases in the atmosphere.
3.2. General trend in East Asia

In general, the concentrations of N-gaseous were relatively higher in Indonesia than those in Taiwan sites. However, the [O₃] and [SO₂] were similar between the two countries (figure 4). Even the concentrations in the forest of Indonesian sites were still higher than the city site in Taiwan (figure 4). Presumably, the higher concentrations in the Indonesian sites could be attributed to the condition of plantations and the locations. The sites in Indonesia belong to a commercial plantation with relatively high activities of harvesting and logging. There is also a pulp mill nearby and busy logging roads that may contribute to the higher amount of those gases in the area. On the other hand, the sites in Taiwan were located in an experimental forest that most activities in the area are recreation and educations. Despite the fact that the concentration of N-derived gases being higher in Indonesia sites, the [O₃] was similar between sites in the two countries.

![Figure 4. The mean value of air pollutant gases concentration in Indonesia and Taiwan.](image)

With the rapid industrialization development in the emerging economies of the Asia region, the atmospheric [SO₂], [NO₃], and [O₃] are rising and causing severe impact on the ecosystems [16]. The WHO established a daily SO₂ guideline of 20 µg m⁻³. About 40% of the cities in Asia had an annual average of SO₂ level equal to or lower than 10 µg m⁻³; however, 24% of the cities had it over the WHO’s level [17]. SO₂ is highly accredited for the acid rain that damages crops, forests, and corresponding ecosystems [17].

Further, the atmospheric [O₃] has increased significantly in recent years, owing to the high NOₓ emission from industrialization and combustion of fossil fuels. In urban areas where NOₓ and hydrocarbon emissions are high, O₃ tends to accumulate rapidly.

Forest plantation has developed rapidly in the past decades to meet the increased demand for pulpwood, sawn timber, plywood, and other wood-based products. During 1981-1990, the areas of forest plantation in the tropics reached a total gross of 43.8 million ha, of which 10 million ha of the area were planted with eucalypts [18]. Acacia, especially *A. mangium*, is one of the most common plantation species in Asia which the largest plantations are in Indonesia and Malaysia [19].

These results implied that N deposition and ozone have not yet become a threat for eucalypts and acacia, whether in the tropic or sub-tropic region. However, the air pollution trend in Asia is escalating, suggesting that the concentration of NOₓ, NO₂, NO, and O₃ in the observed locations might reach a point that the impact on the plant will be more pronounced. Therefore, the responses of those promising afforestation species to the future concentration of N and [O₃] are necessary objects to be examined.
4. Conclusion
Given their association with nitrogen-fixing microbes, the plantation of *A. crassicarpa* might contribute to the higher emission of N-derived gases than the *E. pellita*, which in turn might give rise to the high \([O_3]\) in the area. Furthermore, the age of rotation that correlated with the accumulation of organic matters in the soil might have more significant influence on the N-derived gases emission than the age of the stand that correlated with the open area and the air temperature in the stand.

The \([NO_3]_s\), \([NO_2]_s\), and \([NO]_s\) were higher in Indonesia sites than in Taiwan’s because of the study site-effect in which sites in Indonesia had more intense anthropogenic activities. However, the \([O_3]\) in Indonesia and Taiwan sites were relatively similar, which was still below the threshold level in Indonesia. Therefore, it is crucial to further investigate the responses of eucalypts and acacia to the elevated \([O_3]\) and N deposition.

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