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## Article

# Measuring Ammonia Concentration Distributions with Passive Samplers to Evaluate the Impact of Vehicle Exhaust on a Roadside Environment in Tokyo, Japan

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**Abstract:** Evaluating the impact on roadside environments of NH<sub>3</sub> from vehicle emissions is important for protecting the ecosystem from air pollution by fine particulate matter and nitrogen deposition. This study used passive samplers to measure NH<sub>3</sub> and NO<sub>x</sub> at multiple points near a major road to observe the distribution of these gases in the area. The impact of NH<sub>3</sub> emitted from vehicles on a major road on the environmental concentration of NH<sub>3</sub> at different distances from the roadside was found to be similar to that of NO<sub>x</sub> and NO<sub>2</sub>. The concentration of NH<sub>3</sub> rapidly decreased due to dilution and diffusion within approximately 50 m of the road, and after 100 m the concentration remained almost the same or decreased slowly. Furthermore, CO<sub>2</sub> observations taken in the same period along the roadside and in the background yielded a vehicular emission factor of 4–50 mg/km for NH<sub>3</sub>, which is comparable with previous research. This emission factor level contributes 4–11 ppb to the NH<sub>3</sub> concentrations in roadside air through the dilution and diffusion process. A correlation was found between the emission factors of NH<sub>3</sub> and NO<sub>x</sub> that is different from the trade-off relationship seen when single-vehicle exhaust is measured.

**Keywords:** diffusive sampler; distance attenuation; emission rate; automotives

## 1. Introduction

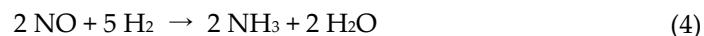
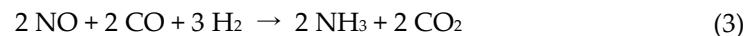
Ammonia (NH<sub>3</sub>) gas in the atmosphere reacts with acidic gases to form ammonium salts, which can increase the concentration of fine particles in the atmosphere [1–4]. In addition, particulate ammonium formed from NH<sub>3</sub> can be transported far from the source [3,5], and semi-volatile ammonium salts (e.g. Equation (1)) can return to NH<sub>3</sub> under specific meteorological and chemical conditions such as concentration, temperature, and humidity [6–9].



NH<sub>3</sub> is highly reactive and is quickly removed, so its concentration decreases rapidly as it moves away from the source [10]. However, even at low concentration levels NH<sub>3</sub> gas has been found to have a direct negative impact on plant communities, and a critical level of 1 µg/m<sup>3</sup> (1.4 ppb at 20°C and 1 atm standard conditions) has been proposed to protect vegetation [11].

It has long been known that NH<sub>3</sub> in the atmosphere is emitted from biological processes in soil, biomass combustion, ammonia-based chemical fertilizers, sewage treatment plants, and the decomposition process of animal waste [3,12]. Since the introduction of three-way catalysts (TWCs) in the 1980s, attention has focused on the contribution of NH<sub>3</sub> emissions from gasoline-powered vehicles to atmospheric NH<sub>3</sub> concentrations [13–18]. TWCs are honeycomb-shaped ceramics coated with rare earth metals such as palladium (Pd), platinum (Pt), and rhodium (Rh). They are designed to control the emission of carbon monoxide (CO), unburned hydrocarbons (HC), and nitrogen oxides

(NOx) from gasoline vehicles through a series of chemical oxidation–reduction reactions that occur on the surface of the catalyst. Thus, a TWC oxidizes CO and unburned HC compounds into carbon dioxide (CO<sub>2</sub>) and water (H<sub>2</sub>O) and reduces NOx such as nitric oxide (NO) to nitrogen gas (N<sub>2</sub>) [19–23]. When NO compounds are excessively reduced beyond the capacity to form N<sub>2</sub> gas on the surface of the TWC, NH<sub>3</sub> is formed, which is mainly related to the reaction between NO and H<sub>2</sub> [13–17,24]. This process usually begins with the water–gas shift reaction between CO and H<sub>2</sub>O, which produces CO<sub>2</sub> and H<sub>2</sub> (Equation (2)), and NH<sub>3</sub> is formed as a result of the reaction between the produced H<sub>2</sub> and NO (Equations (3) and (4)).



Factors that may affect NH<sub>3</sub> formation in TWCs include aggressive driving [25–27], failure to maintain a stoichiometric air–fuel ratio [14,15,26–28], ambient temperature and engine temperature [29], and the age and model year of the TWC [30–34]. The efficiency of the TWC depends on the air–fuel ratio, and there is a narrow operating range around the stoichiometric ratio of air and fuel ( $\lambda = 1$ ) where the TWC operates most efficiently. For this reason, fuel combustion in gasoline vehicles is generally controlled by a lambda sensor to achieve  $\lambda = 1$ , but aggressive driving can change  $\lambda$ . During lean combustion at  $\lambda > 1$ , NOx emissions usually increase, while during fuel-rich conditions of  $\lambda < 1$ , NH<sub>3</sub> emissions increase, so in general, there is a trade-off relationship between NOx and NH<sub>3</sub> emissions [34]. Another known cause of NH<sub>3</sub> emission is NH<sub>3</sub> slip (emission of unreacted NH<sub>3</sub> into the atmosphere due to excessive supply) on urea selective catalytic reduction (SCR) catalysts, which are used to reduce NOx emissions from diesel vehicles [35,36]. Therefore, to understand the complex dynamics of NH<sub>3</sub> concentrations in urban air and provide evidence of the effectiveness of NH<sub>3</sub> mitigation strategies that have been reported in previous studies e.g. [4], it is necessary to consider the impact of vehicle traffic on amounts of NH<sub>3</sub> in the surrounding environment.

Although many papers have been published on monitoring NH<sub>3</sub> in the atmosphere, there are still many issues to be addressed. Because the concentration of NH<sub>3</sub> varies greatly on spatial and temporal scales due to the rapid deposition of this molecule and its reactivity in the atmosphere, high time-resolution measurements using continuous analyzers have been attempted but suffer from measurement uncertainty depending on which analysis technology is used [37]. In addition, there are no standardized quality control/quality assurance procedures for measuring NH<sub>3</sub> and currently no established traceability or international guidelines to support measurements [38]. Passive samplers, which are a classic method for measuring NH<sub>3</sub> [39–45], rely on the diffusion of the target gas onto a surface where it is chemically trapped by an adsorbent. Measurement uncertainty arises both from the preparation of the sampler and in the laboratory analysis, as well as the environmental exposure factors [39–45]. Most studies follow European standard diffusion sampler methodology EN 17346: *Ambient air – Standard methods for the determination of NH<sub>3</sub> concentration using diffusion samplers* [46]. Examples of commercially available passive samplers include the Stream Sampler (Kogawa Seisakusho, Hyogo, Japan) and the Adaptive Low-cost Passive High Absorber (ALPHA) Sampler (Centre for Ecology and Hydrology, Edinburgh, UK). Because passive samplers take samples at a fairly low frequency (from one week to one month), the results obtained are low time-resolution measurements—average values observed over a long period of time—as opposed to those obtained by numerous types of continuous analyzer that respond to rapid concentration changes due to deposition and reactions in the atmosphere. Passive samplers are inexpensive and do not require a power supply, making them suitable for use in both urban and remote areas, where they can be used to measure at fine spatial resolutions and understand spatial variability. Consequently, previous studies using passive samplers have provided a wide range of knowledge about spatial distributions

of air pollutant concentrations [47–49]. However, the components that can be measured using passive sampling are limited to those with high reactivity (such as NH<sub>3</sub>, NO<sub>2</sub>, and O<sub>3</sub>), making it difficult to measure CO<sub>2</sub>, an inert gas that needs to be measured in parallel to evaluate NH<sub>3</sub> emissions from vehicles in real atmospheric environments. Research that uses expensive continuous analyzers to provide CO<sub>2</sub> measurements is limited.

To investigate the effects of vehicle exhaust gases, this study used passive samplers that do not require a battery to determine the spatial distribution of NH<sub>3</sub> concentrations in a roadside environment. Furthermore, we determined the emission levels from vehicle exhaust gases by simultaneously measuring roadside and background CO<sub>2</sub> using a portable, battery-powered, low-cost sensor. The findings of this research are discussed in detail, comparing them with past research. The NH<sub>3</sub> emission factor of several tens of milligrams per kilometer observed by this study was found to contribute to the NH<sub>3</sub> concentration in roadside environments at a level of several parts per billion.

## 2. Materials and Methods

### 2.1. Locations

To measure the planar concentration distribution of NH<sub>3</sub> and NO<sub>x</sub>, passive samplers were installed at 20 sites in the area surrounding a major road. According to Japanese traffic census data [50–52], the traffic volume on Ring Road No. 8 near Nogemachi Park is approximately 52,000 vehicles per day, with a large-vehicle ratio of 17.2% (more details can be found in previous studies [53–55]).

Passive samplers were placed at varying distances from a central observation point (N 35°36'22.60", E 139°38'33.71") referred to as the Noge air quality monitoring station [54], which was located beside a major road in Tokyo (Ring Road No. 8 (National road No. 311)) where it runs between Tamagawa Nogemachi Park and an urban residential area. Samplers were attached to streetlights along the main road, on lampposts in the park area, and on telegraph poles in the residential area, and observations were carried out. This study determined the emission factors (emission levels) from vehicle exhaust gases in a twin-site study by simultaneously measuring roadside and background CO<sub>2</sub> and NH<sub>3</sub>. To additionally measure the background urban atmosphere not directly affected by roadside air, measurements of air pollutants taken at a general monitoring station located approximately 6 km from the roadside of this study were accessed online (measurement station code: 13112010, Setagaya air quality monitoring station, N 35°38'48.12", E 139°39'11.42") [56,57].

### 2.2. Sampling

NH<sub>3</sub> and NO<sub>x</sub> were measured using passive samplers with reference to previous studies that showed the method was consistent with conventional continuous measurement and the Denuder method [39–45,49]. Here, a brief description of the measurement device used in this study is given.

NH<sub>3</sub> concentration was measured using a short-term passive sampler (OG-KN-S, Ogawa & Co., Ltd., Hyogo, Japan) inserted into a rain shelter (OG-SN-S, Ogawa & Co., Ltd., Hyogo, Japan) 2.5 m above the ground. The sampling period was one week. The filter paper (OG-SN-17, Ogawa & Co., Ltd., Hyogo, Japan) was removed from the sampler after collection, extracted with pure water, and the ammonium ion (NH<sub>4</sub><sup>+</sup>) concentration was measured using ion chromatography (Integrion RFIC system, Thermo Fisher Scientific Inc. Waltham, MA, USA). The NH<sub>3</sub> concentration was calculated from this value using a calibration curve prepared separately and then corrected using temperature and humidity data [39–45,49] from the Japan Meteorological Agency's Tokyo Regional Meteorological Office [58] for the study periods.

NO<sub>x</sub> was measured using a long-term passive sampler (OG-KN-S, Ogawa & Co., Ltd., Hyogo, Japan) for measuring NO<sub>2</sub> and NO<sub>x</sub> and filter papers impregnated with reagents, triethanolamine (OG-SN-10) for NO<sub>2</sub> and 2-phenyl-4,4,5,5-tetramethylimidazoline-3-oxide-1-oxyl (OG-SN-11) for NO<sub>x</sub>. The sampling period was one week. The extract with pure water was then colored using

Salzmann's reagent, and the absorbance at a wavelength of 545 nm was measured. The NO<sub>2</sub> and NO<sub>x</sub> concentrations were calculated from these absorbance values using a calibration curve prepared separately and then corrected in the same way as the NH<sub>3</sub> concentrations [39–45,49].

Sampling using the passive samplers was carried out twice on consecutive weeks in each of the four seasons: Autumn-1 (18 to 25 October, 2017), Autumn-2 (25 October to 1 November, 2017), Winter-1 (17 to 24 January, 2018), Winter-2 (24 to 31 January, 2018), Spring-1 (9 to 16 May, 2018), Spring-2 (16 to 23 May, 2018), Summer-1 (18 to 25 July, 2018), and Summer-2 (25 July to 1 August, 2018) to correspond with the monitoring period for PM<sub>2.5</sub> component analysis designated by the Japanese Ministry of the Environment [59].

Low-cost CO<sub>2</sub> sensors (TR-76Ui, T&D Corporation, Matsumoto, Nagano, Japan) were used to roughly measure the effects of vehicle emissions at only two locations: near the roadside where a power supply could be used (Noge Station) and in the background at the general monitoring station in Setagaya. The CO<sub>2</sub> was measured continuously for two weeks with a one-minute resolution during the collection periods by passive sampler.

### 3. Results and Discussion

#### 3.1. Spatial Patterns in Concentrations of NH<sub>3</sub> and NO<sub>x</sub>

The results for all seasons investigated in this study show that the concentrations of NH<sub>3</sub> and NO<sub>x</sub> decreased rapidly up to about 50 m from the road, and beyond 100 m they either remained more or less the same or decreased gradually. The ratio of the highest concentration along the road to the background concentration (Site 20, Setagaya station) shows that the concentrations decreased by 90%  $\pm$  2% for NH<sub>3</sub>, by 61%  $\pm$  9% for NO<sub>2</sub>, and by 80%  $\pm$  6% for NO<sub>x</sub> throughout all seasons.

Several previous studies have shown that the concentrations of NH<sub>3</sub> and NO<sub>2</sub> decrease rapidly as you move away from the roadside. In one study that measured NH<sub>3</sub> in suburban roadside environments with different traffic volumes, a 99% decrease in concentration was observed at a distance of 20 m from the roadside [60]. Another study in suburban roadside environments found a 90% decrease in NH<sub>3</sub> concentration from background levels at a distance of 10 m from the roadside and a 90% decrease in NO<sub>2</sub> concentration at a distance of 15 m from the roadside [61]. When NH<sub>3</sub> and NO<sub>x</sub> were measured in three roadside environments in urban areas, a reduction in concentration of approximately 80% from background levels several hundred meters away from the road was observed at a distance of 50 m from the roadside [62]. Despite the variation in the results found in these studies [60–62], NH<sub>3</sub> concentrations behaved in a similar way to NO<sub>x</sub> and NO<sub>2</sub> concentrations across all the areas under observation, decreasing rapidly at around 50 m from a road, and then either remaining more or less the same or decreasing gradually after 100 m. The reason for the variation in the results of these studies is thought to be the differences in conditions such as the types and numbers of vehicles driving on the roads under observation, the season, and the presence of obstructions [62]. For example, it was found that the NH<sub>3</sub> concentration was 140% higher on average at a distance of 12 km downwind from a major urban area than in the hinterland (upwind of the roadside) of roadside observations in the suburbs [63]. In addition, in urban areas, NH<sub>3</sub> concentrations at roadside locations were observed to be approximately 80% lower than at background locations [64]. Therefore, the observed reduction rate is likely to be affected by the difference between local and background concentrations of pollutants.

This study also found a high correlation between the increase in NH<sub>3</sub> concentration and those of NO<sub>x</sub> and NO<sub>2</sub> (NO<sub>x</sub>:  $r = 0.953 \pm 0.039$ , NO<sub>2</sub>:  $r = 0.909 \pm 0.046$ ). Relative to the regression lines for NH<sub>3</sub> concentration over the entire survey period, the slope of the regression line for NO<sub>x</sub> concentration was  $0.153 \pm 0.024$ , with a relative standard deviation of 16%, and the slope of the regression line for NO<sub>2</sub> concentration was  $0.479 \pm 0.024$ , with a relative standard deviation of 21%. As shown in Equation (1), the generation of NH<sub>3</sub> due to the dissociation of NH<sub>4</sub>NO<sub>3</sub> at high temperatures, such as through the pre-deposition of ammonium ion (NH<sub>4</sub><sup>+</sup>) species together with water (dew, mist, etc.) on grassland or building walls under high relative humidity at night, is an important process that

promotes evaporation in the morning [65,66]. The scatter plots of NH<sub>3</sub>, NO<sub>x</sub>, and NO<sub>2</sub> obtained in this study show a high correlation and a slope that is generally consistent regardless of the season, indicating that the exchange of gases and particles around the roadside and the effects of evaporation from deposited ion species are within the range of variation to be expected in observations taken using passive samplers with low time-resolution measurements.

In line with the findings of previous studies [62], the current study found that the impact of NH<sub>3</sub> emitted from vehicles driving on major roads on environmental concentrations at different distances from the roadside was in the same range as that of NO<sub>x</sub> and NO<sub>2</sub>, and that NH<sub>3</sub> concentrations decreased rapidly due to dilution and diffusion within about 50 m of the road and remained almost unchanged or decreased slowly after 100 m.

### 3.2. Comparison with NH<sub>3</sub> Concentrations in Other Regions and Periods

Table 1 lists the NH<sub>3</sub> concentration ranges found using passive samplers in this and previous studies conducted in other years and various regions.

In roadside observations of vehicle exhaust emissions, NH<sub>3</sub> concentrations in other studies were observed to be in the range of 1–31 ppb [60–62], so the NH<sub>3</sub> concentrations of 4–11 ppb observed at the roadside in 2017–2018 in this study were within the concentration range of previous studies. The variation in NH<sub>3</sub> concentrations at the roadside in other studies depended on the region and length of observation rather than the year of observation. In particular, the highest NH<sub>3</sub> concentrations (32–129 ppb) were reported for measurements taken in road tunnels, where there are no sources other than vehicles and NH<sub>3</sub> concentration is not diluted. It is known that NH<sub>3</sub> is emitted from gasoline, diesel, and liquefied petroleum gas vehicles, and some of the nitrogen oxides emitted from vehicles are reduced to NH<sub>3</sub> by the TWC installed in the vehicle and released into the atmosphere [13–18]. Vehicle exhaust is recognized as one of the main sources of NH<sub>3</sub> in urban areas [80,81], and reports have shown that NH<sub>3</sub> emissions are increasing not only in areas with high vehicle density but also in agricultural areas [75,82]. Regarding background levels, Table 1 shows that the NH<sub>3</sub> concentration along the roadside tended to be high as that in one of the industrial city areas (6–11 ppb). The background NH<sub>3</sub> concentration in urban areas is reported to be 1–85 ppb, and in suburban and rural areas 1–78 ppb. The NH<sub>3</sub> concentration of 1–5 ppb observed in the urban background in this study was therefore within the concentration range of previous studies. The comparison of data in Table 1 also shows that the variation in NH<sub>3</sub> concentration in urban areas (1–85 ppb) was wider than the variation in NH<sub>3</sub> concentration in roadside environments (1–31 ppb).

In the locations shown in Table 1, livestock areas and sewage treatment plants were the sources of the highest concentrations of NH<sub>3</sub> (266–8455 ppb), and a wide range of concentrations were observed in these environments. NH<sub>3</sub> emissions have also been reported from landfill sites and incineration facilities that process general waste generated in residential areas [68], from electronic cigarettes [83], and from plant combustion in urban areas [84]. To understand the various characteristics of NH<sub>3</sub> emissions, detailed data on concentrations and their sources in each case is important. A potential source of NH<sub>3</sub> that has attracted attention in recent years is a technique for co-firing NH<sub>3</sub> to reduce CO<sub>2</sub> emissions from internal combustion engines in vehicles and coal-fired power plants [48]. Additionally, NH<sub>3</sub> emissions due to NH<sub>3</sub> slip are an example of NH<sub>3</sub> combustion [85,86]. Therefore, although it should be emphasized that the increase in NH<sub>3</sub> concentrations found in the present study between the background and the roadside is due to vehicle exhausts, it should also be noted that high NH<sub>3</sub> concentrations of non-motor vehicle origin may also be important.

**Table 1.** NH<sub>3</sub> concentration ranges measured in studies using passive samplers at various locations and in different periods

Location	Region	Period	NH <sub>3</sub> [ppb] <sup>*1</sup>	Reference
Roadside	Tokyo, Japan	2017–2018	4–11	This Study
Roadside	Saitama, Japan	2005–2007	6–31	[62]

Roadside	London, UK	2006–2019	4–7	[67]
Roadside	Barcelona, Spain	2010–2011	1–25	[68]
Roadside	Gyeonggi, Korea	2020–2021	12–20	[69]
Roadside	2 sites, Korea	2022	8–26	[49]
Road tunnel	Beijing, China	2014–2015	32–129	[70]
Urban	Tokyo, Japan	2017–2018	1–5	This Study
Urban	2 sites, North America	2003–2014	1–4	[71]
Urban	Xi'an, China	2006–2007	1–52	[72]
Urban	Beijing, China	2008–2010	1–85	[73]
Urban	Barcelona, Spain	2010–2011	6–55	[68]
Urban	13 sites, China	2015–2016	4–41	[48]
Urban	New York, USA	2016–2017	0.1–5	[74]
Urban	Beijing, China	2019	13–32	[75]
Urban	Gyeonggi, Korea	2020–2021	4–23	[69]
Urban	3 sites, Korea	2022	2–24	[49]
Urban background	Gyeonggi, Korea	2020–2021	2–5	[69]
Urban and rural	Asia, Africa, and South America	1999–2001	1–20	[47]
Suburban	Edinburgh, UK	2006–2019	1–3	[67]
Suburban	Xi'an, China	2006–2007	1–78	[72]
Suburban	Colorado, USA	2010–2015	3–15	[76]
Rural	11 sites, North America	2003–2015	0.2–6	[48]
Rural	Beijing, China	2007–2010	1–43	[73]
Rural	Colorado, USA	2010–2015	1–8	[76]
Rural, remote	40 sites, China	2015–2016	0.4–25	[48]
Rural	New York, USA	2016–2017	0.2–5	[74]
Rural	Jeongeup, Korea	2019–2020	11–38	[77]
Rural	5 sites, Korea	2022	0.8–6	[49]
Industrial	Gyeonggi, Korea	2020–2021	6–11	[69]
Industrial	10 sites, Korea	2022	4–87	[49]
Agricultural	North Carolina, USA	2003–2004	5–21	[78]
Agricultural	Colorado, USA	2010–2015	5–104	[76]
Agricultural	Navarre, Spain	2013–2015	7–79	[79]
Agricultural	Nanjing, China	2015–2016	7–57	[48]
Agricultural	8 sites, Korea	2022	6–35	[49]
Livestock	Beijing, China	2014–2015	670–2129	[70]
Livestock	Gyeonggi, Korea	2020–2021	32–96	[69]
Livestock	7 sites, Korea	2022	15–266	[49]
Waste plants	Beijing, China	2014–2015	186–8455	[70]

\*1: Reported concentrations of mass per volume ( $\mu\text{g}/\text{m}^3$ ) were converted to ppb under standard conditions of 20°C and 1 atm.

### 3.3. $\text{NH}_3$ Emission Factors

The  $\text{NH}_3$  emission factors found in this study are based on low-cost sensor observations of  $\text{NH}_3$  and  $\text{NO}_x$  using passive sampling and  $\text{CO}_2$  sensors. A comparison with  $\text{NH}_3$  emission factors based

on different approaches that require expensive and diverse equipment that can focus on different targets helps to clarify the bulk emission factors from vehicles in a given area. Therefore, Table 2 compares the NH<sub>3</sub> emission factors based on distance and fuel measured in this study, as calculated from the NH<sub>3</sub> to CO<sub>2</sub> conversion ratio, with the results of previous studies obtained from dynamometers, in tunnels, by remote sensing, and in on-road experiments.

**Table 2.** NH<sub>3</sub> emission factors for vehicles in other regions and periods

Location or Target	Measurement Device	Year	NH <sub>3</sub> Emissions [mg/km/vehicle]	Reference
Roadside, Tokyo, Japan	Passive sampler	2017–2018	4–50	This Study
Urban, Tokyo, Japan	Semi-continuous analyzer	2017	3.7–32	[87]
Van Nuys Tunnel, California, USA	Filter pack	1993	61	[88]
Caldecott Tunnel, California, USA	Denuder	1999	46–52	[89]
Gurbrist Tunnel, Switzerland	Continuous analyzer	2002	26–35	[90]
Jânio Quadros Tunnel, São Paulo, Brazil	Impinger	2011	20–64	[91]
Tunnel, Guangzhou, China	Semi-continuous analyzer	2013	216–119	[92]
Handan Tunnel, Shanghai, China	Passive	2014	23–52	[18]
On-road, California, USA	Remote sensing	1996	86–102	[93]
Gasoline vehicle	Chassis dynamometer	2002	2–110	[94]
Gasoline vehicle	Chassis dynamometer	2004	1–31	[95]
Gasoline vehicle	Chassis dynamometer	2006	3–256	[96]
Gasoline vehicle	Chassis dynamometer	2008	3–28	[27]
Gasoline vehicle	Chassis dynamometer	2014	4–70	[97]
Gasoline vehicle	Chassis dynamometer	2017	2–132	[98]
Gasoline vehicle	Chassis dynamometer	2018	5–53	[32]
Gasoline vehicle	Chassis dynamometer	2022	1–53	[99]
Gasoline vehicle	On road	2020	1–53	[100]
Diesel vehicle	On road	2020	1–32	[100]
Compressed natural gas vehicle	On road	2020	38–90	[100]
Gasoline vehicle	Emission inventory	2018	29–104	[101]
Diesel vehicle	Emission inventory	2018	0.6–1.7	[101]
Gasoline vehicle, Japan	Emission inventory	2020	0.1–95	[102]

Emission factors are also expressed as mg/kg-fuel normalized per CO<sub>2</sub> emission and mg/km normalized per driving distance. Emission factors used to create emission inventories, which are usually used as data to analyze the effectiveness of the vehicle emissions type certification process and emission regulations, express emissions in distance-based factors such as grams per kilometer or per mile [103].

Methods for estimating emission factors from atmospheric observations have been summarized in a systematic literature review [104]. In this study, emission factors were estimated by Equation (5), which is based on that used for twin-site studies (i.e., simultaneous measurement of the roadside and background) [104]:

$$\text{Emission Factor} = \frac{([NH_3]_{road} - [NH_3]_{background})}{([CO_2]_{road} - [CO_2]_{background})} \times W_{CO_2} \times FC \quad (5)$$

where [NH<sub>3</sub>]<sub>road</sub> and [NH<sub>3</sub>]<sub>background</sub> are the NH<sub>3</sub> concentrations at road (Site 0) and background (Site 20) (mg/m<sup>3</sup>); [CO<sub>2</sub>]<sub>road</sub> and [CO<sub>2</sub>]<sub>background</sub> are the CO<sub>2</sub> concentrations at road (Site 0) and background

(Site 20) (kg-CO<sub>2</sub>/m<sup>3</sup>); W<sub>CO<sub>2</sub></sub> is the content of carbon in fuel (kg-CO<sub>2</sub>/L), taking into account the proportion of gasoline and diesel vehicles; and FC is distance-based fuel consumption volume (L/km), taking into account the proportion of gasoline and diesel vehicles.

W<sub>CO<sub>2</sub></sub> is estimated by Equation (6):

$$W_{CO_2} = W_{CO_2}(\text{Gasoline}) \times \frac{100 - P_{diesel}}{100} + W_{CO_2}(\text{Diesel}) \times \frac{P_{diesel}}{100} \quad (6)$$

where W<sub>CO<sub>2</sub></sub> (Gasoline) is the content of carbon in gasoline fuel (2.32 kg-CO<sub>2</sub>/L) and W<sub>CO<sub>2</sub></sub> (Diesel) is the content of carbon in diesel fuel (2.49 kg-CO<sub>2</sub>/L), and P<sub>diesel</sub> is the proportion of diesel vehicles (heavy-duty vehicles) (17.2%).

FC is estimated by Equation (7):

$$FC = FC(\text{Gasoline}) \times \frac{100 - P_{diesel}}{100} + FC(\text{Diesel}) \times \frac{P_{diesel}}{100} \quad (7)$$

where FC (Gasoline) is the statistical estimate value for a gasoline vehicle (0.085 L/km) [105] and FC (Diesel) is the statistical estimate value for a diesel vehicle (0.220 L/km) [105].

Distance-based NH<sub>3</sub> emission factors for individual vehicles obtained in the past—from dynamometer experiments, in vehicle tracking studies on roads, and in emission inventories—vary widely from 0.1 to 256 mg/km (Table 2). In general, vehicles driven at aggressive driving cycles usually have high NH<sub>3</sub> emission factors [97,98,100], and the aging of the TWC and of the vehicle [31–33] also contribute to the large variation. The measurements taken in previous studies, both in road tunnels and by remote sensing, also showed a wide variation, ranging from 3.7 to 119 mg/km. The results of this study fell within the range of the wide variation in the results of previous experiments and in measurements taken under actual atmospheric conditions. In particular, the results of this study were generally consistent with the results of measurements taken at different locations and using different methods in urban atmospheres.

Measurements of NH<sub>3</sub> taken in road tunnels, on roads, and in urban air cover all types of vehicles, both gasoline and diesel vehicles. However, I view the impact of diesel vehicles on NH<sub>3</sub> emissions to be limited in this study. This is because heavy vehicles account for approximately 17.2% of the vehicles passing along the roadside in this study. In Japan, the introduction of SCR systems using urea for heavy trucks and other vehicles began in the mid-2000s [106]. SCR systems produce more NH<sub>3</sub> [35,36], and there is a possibility that they increase the emission factor of NH<sub>3</sub> from diesel vehicles. At the time this research was conducted in 2017, the number of diesel vehicles (heavy vehicles) sold in Japan with SCR systems installed was 634,000 vehicles [106], which is about 4% of the total number of vehicles owned in Japan (14,652,000 vehicles) [107]. Vehicles without SCR systems generally have low NH<sub>3</sub> emissions [32] and have little effect on the results of emissions, so in this study the impact of diesel vehicles on NH<sub>3</sub> emissions would be limited.

It is reported that NH<sub>3</sub> emissions from urban soil in green spaces are highly likely to have contributed spikes in atmospheric NH<sub>3</sub> that occur after the morning rush hour [108]. The timing of the morning peak after the morning rush hour, as measured by a device with a one-hour time resolution, is consistent with the results of a previous study [108]. NH<sub>3</sub> emissions from soil are also partially dependent on the ambient temperature. However, a study that estimated the NH<sub>3</sub> emission coefficient by observing the NH<sub>3</sub> concentration in the general atmosphere of Tokyo suggested that the impact of emissions from urban soil in green spaces is small, as although emissions from green space soil may be a source of NH<sub>3</sub> in urban areas, they are insufficient to cause an NH<sub>3</sub> peak equivalent to or greater than that of annual vehicle emissions [87]. Therefore, although the NH<sub>3</sub> emission factor estimated in this study includes emissions from urban soil in green spaces, it is within the range of variation in observations taken using passive samplers at low time-resolution measurements.

This study has found some interesting results, including a correlation between the emission factors for NH<sub>3</sub> and NO<sub>x</sub> calculated from measurements taken in the atmosphere. In this study, the use of passive samplers with low time-resolution measurements gave a bulk measurement (averaged

result) in which the numbers of gasoline and diesel vehicles were mixed in a specific ratio, whereas previous studies have observed clear differences (both trade-offs and correlations) between the NH<sub>3</sub> and NO<sub>x</sub> emission factors measured from gasoline and diesel vehicles individually [91]. Therefore, some explanation is needed of the factors involved in differences between the measured values observed in the atmosphere in this study and the measured values for individual vehicles. First, diesel vehicles emit much less NH<sub>3</sub> than gasoline vehicles but emit more NO<sub>x</sub>. In the case of gasoline vehicles, the NO<sub>x</sub> emitted from the engine is minimized by the reduction reaction in the TWC. Therefore, there is a trade-off relationship between NO<sub>x</sub> and NH<sub>3</sub> emissions, meaning that it is difficult to completely suppress both NO<sub>x</sub> and NH<sub>3</sub> emissions from certain vehicle types [34,91]. Second, it has also been reported that as the driving distance of gasoline vehicles increases, both NH<sub>3</sub> and NO<sub>x</sub> emissions increase [34,91]. Under certain conditions in TWCs, excessive reduction of NO<sub>x</sub> occurs, so the target product of N<sub>2</sub> is not obtained, and NH<sub>3</sub> is produced as an unintended byproduct (Equations (3) and (4)). The efficiency of a TWC depends on the air-fuel ratio, and combustion is controlled to ensure that the TWC operates within a narrow operating range where it is most efficient (air-fuel stoichiometric ratio of  $\lambda = 1$ ). When components such as the catalyst surface and lambda sensor deteriorate over time, the air-fuel ratio may become difficult to control. In addition, when the catalyst deteriorates over time, the catalytic activity and oxygen storage capacity decrease, so the reduction efficiency decreases because there is insufficient O<sub>2</sub> for the redox reaction [34]. In Japan, the average age of passenger cars in 2017 was 12.91 years [109], and further research into emissions from vehicles in use will help to clarify the impact of reductions in TWC efficiency. Therefore, we consider that a combination of these effects is present in the correlation between the emission coefficients of NH<sub>3</sub> and NO<sub>x</sub> found in the atmospheric observations at low time-resolution measurements taken in this study.

The above comparison of results from previous studies and those found in this study emphasizes the need for further research on different parameters affecting NH<sub>3</sub> concentration to help estimate NH<sub>3</sub> emission factors for large volumes of vehicle activity.

#### 4. Conclusions

This study used passive samplers to observe NH<sub>3</sub> and NO<sub>x</sub> and clarify the planar distribution around the road; that is, the attenuation of concentration with distance from the road. The research design highlights the potential for low-cost, multi-point observation as an evaluation tool that can contribute to improving the air quality of roadside environments. In addition, by adding CO<sub>2</sub> measurements using a low-cost sensor, it was shown that the values were within the range of reasonable vehicle emission factors, compared to the considerably large and varied measurement values of previous studies. A correlation found between the NH<sub>3</sub> and NO<sub>x</sub> emission factors obtained in this study were contrary to the trade-off relationship between NH<sub>3</sub> and NO<sub>x</sub> measured for individual vehicles in previous studies. The low time-resolution measurement method used in this study means that the average values represent the combined effects of gasoline and diesel vehicles, which both affect the emission factors in different ways. Even if regulations to mitigate the effects of NH<sub>3</sub> concentration are enforced on new and used vehicles, it will take a considerable amount of time for the effects of the regulations to be seen. For this reason, it will also be important to take temporary measures such as updating road structures (for example, by installing soundproof walls and greening), urban structures (for example, by modifying urban structures to separate traffic networks for people and vehicles to reduce congestion), and traffic regulations. The research design used in this study demonstrates the potential for low-cost, multi-point observation as an evaluation tool that can contribute to improving the air quality of roadside environments.

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