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Impact of NH_3 Emissions on Particulate Matter Pollution in South Korea: A Case Study of the Seoul Metropolitan Area

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Abstract: We analyzed the multi-year relationship between particulate matter (PM_{10} and $PM_{2.5}$) concentrations and possible precursors including NO_2, SO_2, and NH_3 based on local observations over the Seoul Metropolitan Area (SMA) from 2015 to 2017. Surface NH_3 concentrations were obtained from Cross-track Infrared Sounder (CrIS) retrievals, while other pollutants were observed at 142 ground sites. We found that NH_3 had the highest correlation with $PM_{2.5}$ ($R = 0.51$) compared to other precursors such as NO_2 and SO_2 (R of 0.16 and 0.14, respectively). The correlations indicate that NH_3 emissions are likely a limiting factor in controlling $PM_{2.5}$ over the SMA in a high-NOx environment. This implies that the current Korean policy urgently requires tools for controlling local NH_3 emissions from the livestock industry (for example, from hog manure). These findings provide the first satellite-based trace gas evidence that implementing an NH_3 control strategy could play a key role in improving air quality in the SMA.

Keywords: ammonia; particulate pollution; Seoul Metropolitan Area; Korea; CrIS

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1. Introduction

Particulate matter (PM) pollution in South Korea has improved since the 1990s due to active local and nationwide policies, including those specific to the Seoul Metropolitan Area (SMA), but concentrations have not noticeably changed since 2012 [1,2]. There are many factors affecting PM pollution in South Korea, including (1) meteorological conditions and thermodynamics between atmospheric compositions such as nitrate, sulfate, and ammonia [1,3–7] and (2) uncertainty regarding the national emissions inventories reported by the Korea-United States Air Quality Study (underestimations of local volatile organic compounds (VOCs) in Korea) [8] and the underestimation of national NOx emissions by Goldberg et al. [9]. $PM_{2.5}$ formed from ammonium nitrate (NH_3NO_3) is an important contributor to high PM concentrations in Korea and China when atmospheric conditions are stagnant [10–15]. Recent studies have found that ammonium nitrate is the dominant form of $PM_{2.5}$ in Korea; the proportion of ammonium nitrate detected in the atmosphere tends to double (nearly 38% from NH_4 and NO_3, combined) during high-PM episodes [16,17]. The portion of NH_4 in the composition of PM_1 was 14%, estimated from the in situ gas and aerosol observations from a NASA DC-8 during the NASA-NIER KORUS-AQ (Korea-United States Air Quality) campaign ran from May to June of 2016 [18].

Efforts made by the Korean Government to control PM concentrations include reducing NOx emissions from coal power plants, motor vehicles and the industrial sector [19]. The Korean National Assembly has also enacted a law to strongly enforce PM pollution policies [20]. SOx emissions are considered local control issues, such as shipping near national harbors [21], or influenced by transboundary transport (i.e., Chinese emissions) [15,22]. Emissions estimates of volatile organic compounds (VOCs) from industry came to approximately one million tons per year in 2015 [23], but this figure was found to be an underestimate due to limited emissions monitoring around industrial areas [8,13,21]. Ammonia emissions are also a contributing factor to PM formation [15,24–26] and the majority of these emissions in Korea are from the domestic livestock industry (>75% [23,27]), but as of yet there is no specific mitigation target or policy in place.

The Korean Ministry of Environment (KMOE) is currently devising a basic plan for regional air quality management, since the characteristics of PM pollution depend on geographic location and local emissions sources. Specifically, regional atmospheric chemical conditions (e.g., the HOx–NOy relationship) need to be better understood considering the shorter lifetime of atmospheric chemical species such as ammonia and VOCs.

Here, we tried to identify the main contributing factors to PM formation in South Korea with ground measurements of nitrogen dioxide (NO_2), sulfur dioxide (SO_2), carbon monoxide (CO), PM_{10}, and $PM_{2.5}$ obtained nationwide from 2015 to 2017. In addition, we used the National Oceanic and Atmospheric Administration's (NOAA) Cross-track Infrared Sounder (CrIS) NH_3 surface concentration data, since there are few NH_3 ground measurements in South Korea. We used multi-year correlations between chemical species to help identify the main species or emission sources contributing to local PM formation. We focused on the Seoul Metropolitan Area (SMA) to determine the regional contributing species, as dense ground measurements in the area support an understanding of the regional chemical environment.

2. Data and Methodology

2.1. Ground Measurements

In this study, data on air pollutants including NO_2, SO_2, CO, PM_{10} and $PM_{2.5}$ collected at 142 national air monitoring stations were used (more than 75% of data available on an annual basis in SMA [6]).

Air pollutants such as SO_2 and CO were measured using official test methods established by the Environmental Policy Act of Korea [28]. SO_2 was measured using the pulsed ultraviolet (UV) fluorescence method, CO was measured via the non-dispersive infrared method, NO_2 was measured using chemiluminescence analyses with a molybdenum convector and PM_{10} was measured via β-ray absorption [6]. Quality control and quality assessment of instruments were regularly conducted in accordance with the Environmental Examination and Inspection Act [28]. NO_2 should be considered as the sum of NO_2 and NO_z, as the molybdenum convertor causes positive interference with NO_z [29]. The detection limits of SO_2, CO, NO_2, and PM are 0.1, 50, 0.1 ppb and 5 $\mu g/m^3$, respectively [28,29]. The measurements from the 142 stations in the SMA were sampled throughout the experimental period (more than 75% of data available on an annual basis) and used to perform correlation analyses.

2.2. Cross-Track Infrared Sounder Data

A cross-track infrared sounder (CrIS) is a satellite instrument with a Fourier-transform spectrometer onboard the Suomi National Polar-orbiting Partnership (NPP) satellite, which is part of the Joint Polar Satellite System (JPSS) program [30], launched in 2011. CrIS is in a sun-synchronous orbit, with mean local overpass times of 13:30 (ascending node) and 01:30 (descending node) [31–34]. CrIS has three bands in the infrared region (645–1095, 1210–1750, and 2155–2550 cm^{-1}) with a 2200 km swath width and sets of three-by-three circular footprints, each approximately 14 km at the nadir [31–33]. The maximum sensitivity of CrIS NH_3 is in the range of 900–750 hPa and the minimum detection limit near the surface is typically ~1 ppbv [31].

We used surface NH_3 concentration data over South Korea during the period from 2015 to 2017 from CrIS Fast Physical Retrieval (CFPR) version 1.5 [31,34]. We used daytime measurements and removed low-quality data or outliers by applying the standard of quality flag (less than 4), degrees-of-freedom-of-signal (less than or equal to 0.1) and χ^2 (greater than or equal to 0.5). These criteria were selected based on stronger correlation with two Korean NH_3 ground based sites (Imsil, in the southwestern tip of Korea, and Kangwha, on the western coast near the SMA.) with a range of $0.5 < R < 0.7$. The comparison for these local point source measurements with the regional satellite pixel observations showed that the satellite had systemic underestimation by a factor of 2.5 in this location. The range of the long-term (2000–2019) annual mean of ground ammonia measurements in Korea from the Acid Deposition Monitoring Network in East Asia (EANET) sites is 2.7~6.5 ppbv (https://monitoring.eanet.asia/document/public/index, accessed on 10 June 2020).

In total, 1512 CrIS data pixels were used from 2015 to 2017 (42 local samples per month); the retrieval method and data version 1.5 product have been described previously [31,34]. As there is limited independent vertical information [34], the surface and column CrIS NH_3 products are highly correlated and show similar spatial variations over South Korea (not shown).

3. Results

We calculated the three-year correlations among the annual mean of PM_{10}, $PM_{2.5}$, NH_3, NO_2, and SO_2 concentrations over the SMA from 2015 to 2017. We sampled all datasets on an annual basis with a horizontal resolution of 15 km to adjust for the footprint of the nadir CrIS (14 km), producing ~100 samples of the chemical species for the correlational analysis (the sample sizes and locations are shown in Figure 1).

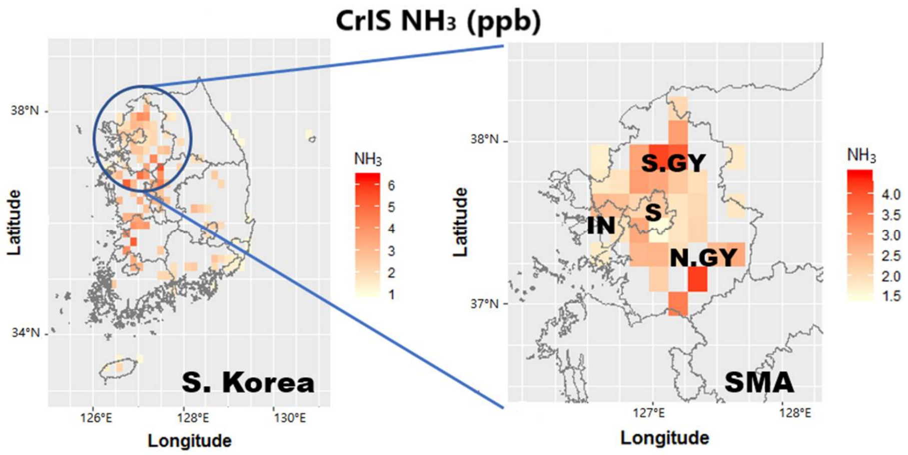

Figure 1. The average concentrations of CrIS surface level NH_3 concentration data (unit: ppb) over South Korea (2015–2017) (**left**) and the data over Seoul Metropolitan Area (SMA), which consists of Seoul (denoted as "S"), Incheon (denoted as "IN"), the northern Gyeonggi area (denoted as "N.GY"), and the southern Gyeonggi area (denoted as "S.GY") are shown (**right**). The grid with less than 1.5 ppbv were masked out.

The relatively higher CrIS NH_3 concentrations over the northern and southern parts of the SMA ("N.GY" and "S.GY" in Figure 1) and the short lifetime of ammonia reveal that there are likely substantial ammonia emissions from the regional livestock industry (around 5000 tons per year; Pocheon in N.GY and Anseong and Icheon in S.GY; NIER, 2018b) (Figure 2) in these regions.

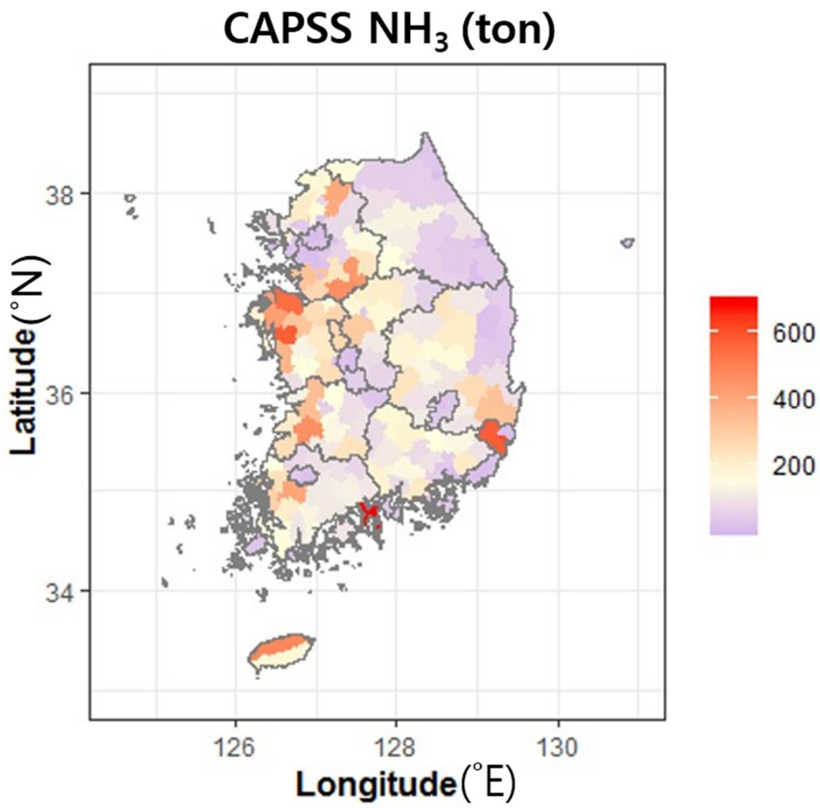

Figure 2. Annual ammonia emissions at local administrative levels in 2015, based on national emissions inventory (CAPSS, NIER, 2018).

Table 1 shows that the NH_3 concentrations have the strongest correlation with $PM_{2.5}$ (R = 0.51) compared to other major PM precursors such as NO_2 or SO_2 (at R = 0.16 and 0.14, respectively). The correlation between NH_3 and $PM_{2.5}$ over the SMA, a relatively high-NOx chemical environment [6,9] (around 25 ppbv, Table 2), was higher than that of all national-level observations (R = 0.31, not in the table). The correlation between NH_3 and $PM_{2.5}$ was particularly high (R = 0.72) in 2016, when the $PM_{2.5}$ levels over SMA were highest over the sample period. The correlation between NH_3 and $PM_{2.5}$ was stronger than the correlation between NH_3 and PM_{10}, implying that local atmospheric ammonia participated in secondary formation.

Table 1. Correlation coefficients between the annual means of PM_{10}, $PM_{2.5}$, NH_3, NO_2, CO, and SO_2 as measured at the national stations and by CrIS retrievals (for NH_3) over the Seoul Metropolitan Area (SMA) between 2015 and 2017. The correlation coefficients in the parentheses indicate correlations excluding Seoul and Incheon ("S" and "IN" in Figure 1), which have greater emissions from transportation and industry. The horizontal scale was set at 15 km according to CrIS footprints.

	PM_{10}	$PM_{2.5}$	NH_3	NO_2	SO_2
PM_{10}	1				
$PM_{2.5}$	0.66 (0.60) ***	1			
NH_3	0.32 (0.35) *	0.51 (0.59) ***	1		
NO_2	0.23 (0.011)	0.16 (0.09)	−0.022 (0.20)	1	
SO_2	0.17 (0.29)	0.14 (0.17)	−0.015 (0.14)	0.38 (0.33) **	1

Note: *** ($p < 0.000$), ** ($p < 0.001$), * ($p < 0.01$).

Table 2. Average concentrations of air pollutants including PM_{10}, $PM_{2.5}$, NH_3, NO_2, and SO_2 in Seoul Metropolitan Area (SMA), South Korea from 2015 to 2017.

Pollutants (Units)	SMA
PM_{10} (μg/m^3)	51 ± 4
$PM_{2.5}$ (μg/m^3)	26 ± 3
NH_3 (ppb)	2.26 ± 0.7
NO_2 (ppb)	25 ± 9
SO_2 (ppb)	5 ± 1

Note: Average ± SD.

However, caution should be exercised when explaining the contribution of ammonia to $PM_{2.5}$ over the entire SMA, owing to the complexity of diverse emissions and the atmospheric chemical environment, with emissions from transportation and industry over the cities of Seoul and Incheon ("S" and "IN" in Figure 1). Furthermore, there are studies demonstrating that the atmospheric conditions of Seoul have been ammonium-rich for two decades now, contributing to the increase in NH_3NO_3 via homogenous and heterogeneous reactions [14,35].

We calculated an additional correlation over the SMA excluding Seoul and Incheon; NH_3 concentrations produced an even stronger correlation with $PM_{2.5}$ (R = 0.59, Table 1). These results provide the clearest evidence yet of the contributions of rural livestock ammonia emissions to $PM_{2.5}$ formation in the vicinity of the SMA (i.e., "N.GY" and "S.GY" in Figure 1), as this region includes cities such as Pochen, Anseong and Icheon, which are home to large livestock operations [8].

Figure 3 shows the average concentrations of PM_{10}, $PM_{2.5}$, NO_2, and NH_3 over the SMA from 2015 to 2017 at a 15 km horizontal resolution. The average NO_2 concentrations over the SMA are consistently higher than 20 ppbv, with the highest over central Seoul (nearly 40 ppbv; Figure 4). The NO_2 concentrations and the corresponding national NOx emission amounts over the SMA are reportedly underestimated [9], implying that the SMA is host to NOx-rich conditions. While the spatial distributions of PM_{10} and $PM_{2.5}$ are different from that of NO_2, the PM distributions are more similar to those in the CrIS NH_3 data (Figure 3). In particular, the northern and southern peaks of NH_3 levels ("N.GY" and "S.GY" in Figure 1) explain the higher $PM_{2.5}$ and PM_{10} concentrations (Figure 3), which strongly suggests that local ammonia emissions from the livestock industry could be a limiting factor in controlling regional $PM_{2.5}$ secondary formation.

PM concentrations over the SMA in Korea peak from December to March, when the impact of the continental emissions is stronger, and gradually decrease from April to September (Figure 5). On the other hand, the degree of secondary PM formation during winter decreases until April but rebounds from April to July, as reflected in $PM_{2.5}/PM_{10}$ ratios (Figure 5). Ammonia concentrations in the SMA are higher between April and June, and this seasonal peak of ammonia is likely associated with higher secondary $PM_{2.5}$ formation in the SMA, which is supported by the highest correlation between $PM_{2.5}/PM_{10}$ and NH_3 in May and June (R^2 = 0.43, Figure 6), when the secondary formation of PM by local ammonia emissions such as livestock waste is relatively important. This relationship can be also explained by another study that found that the greater partitioning of nitrate (NO_3^-) with higher ammonia conditions (and a higher PH) during the warm/dry season (typically May–June) in Korea can produce greater local $PM_{2.5}$ formation [7].

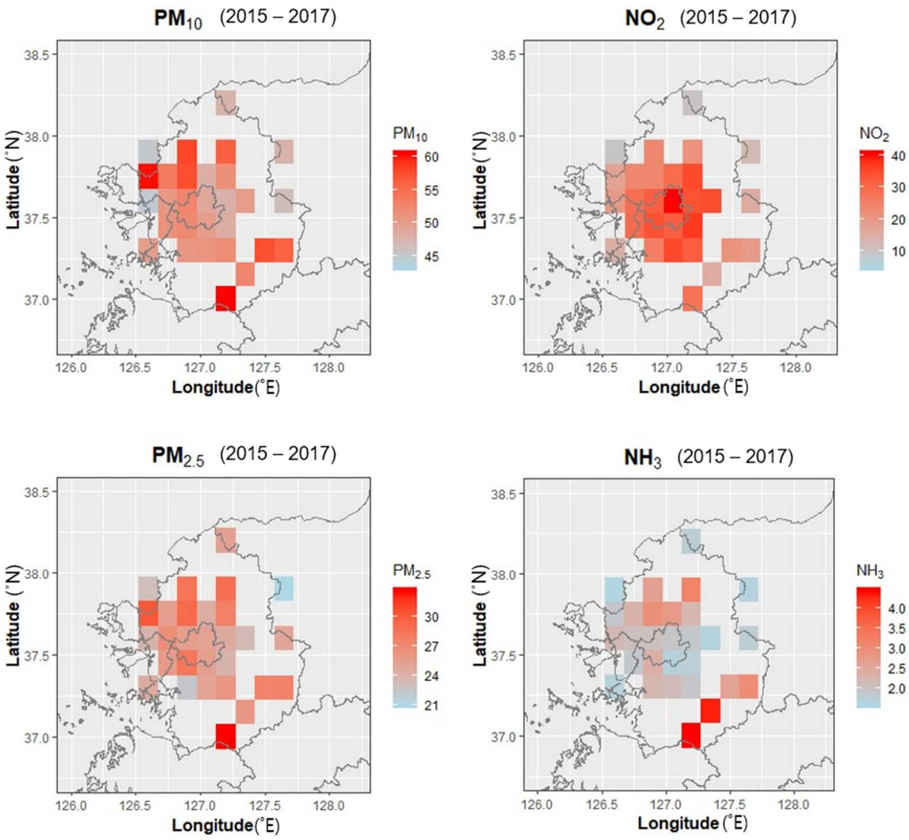

Figure 3. Average concentrations of PM_{10}, $PM_{2.5}$, NO_2, and NH_3 in the Seoul Metropolitan Area (SMA) from 2015 to 2017 (units: PM_{10} and $PM_{2.5}$ ($\mu g/m^3$); NO_2 and NH_3 (ppbv)). The colored area indicates an area where all the chemical species' measurements are coincidently available.

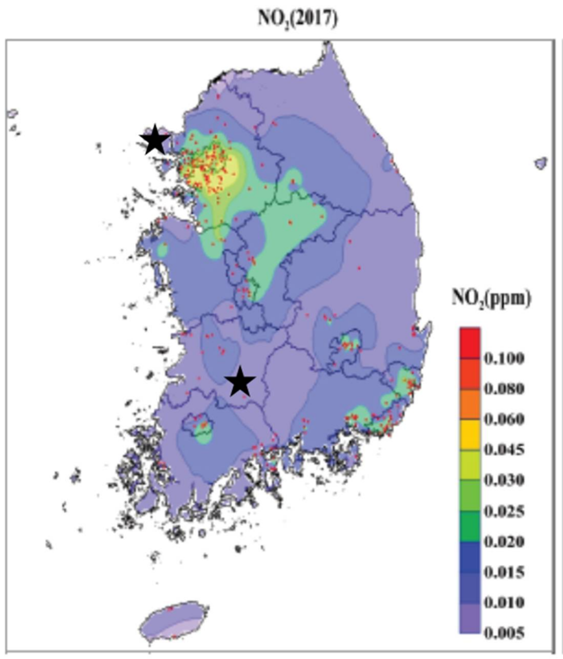

Figure 4. The contour plot of annual mean NO_2 concentrations over South Korea made only from ground measurements in 2017 (adapted from NIER, 2018). The ground measurements of ammonia at Kangwha (denoted as the northernmost black star) and at Imsil (denoted as the southernmost black star) for validating the CrIS satellite retrievals are shown.

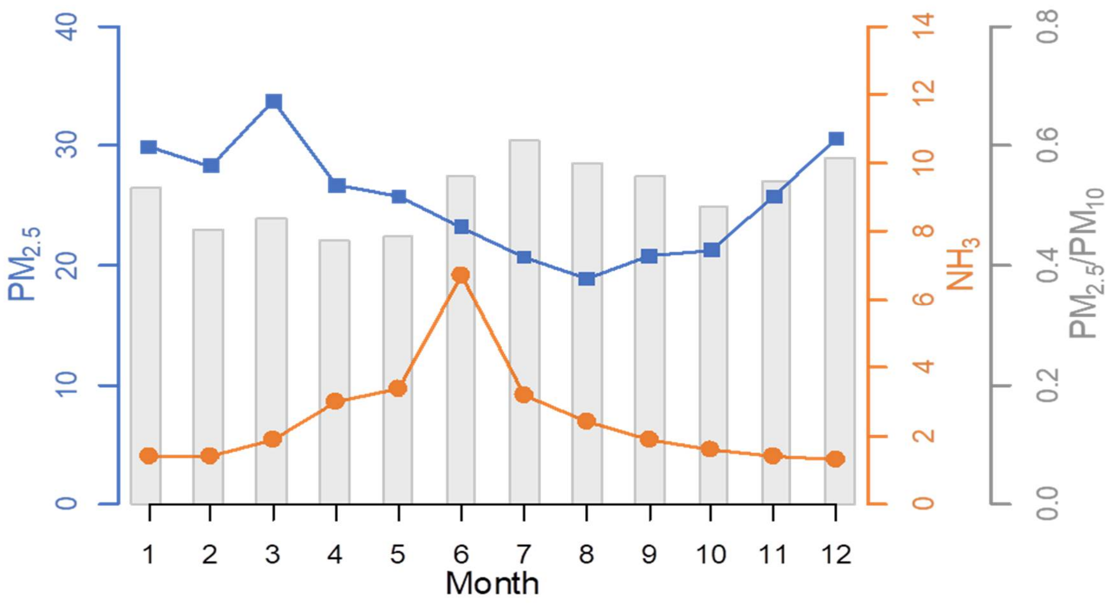

Figure 5. Three-year monthly variations of $PM_{2.5}$ (blue line), CrIS NH_3 (orange line), and $PM_{2.5}/PM_{10}$ ratio (%, gray bars) over the SMA. These data were averaged from the measurements estimated with 15 km-grids (shown in Figure 3).

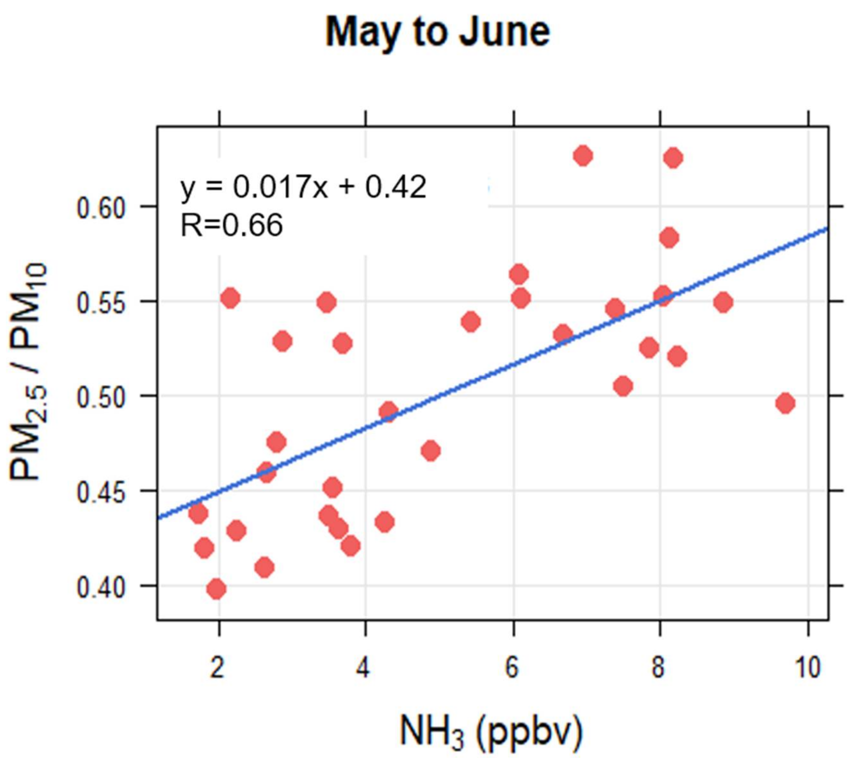

Figure 6. Scatter plots representing the correlation between $PM_{2.5}/PM_{10}$ ratio and CrIS NH_3 over the SMA from May to June, when secondary PM formation was increasing. The data were prepared from monthly means at a 15 km resolution (shown in Figure 3).

4. Discussion

We analyzed the multi-year relationship of particulate matter (PM) concentrations with precursors such as NO_2, SO_2 and NH_3 over the Seoul Metropolitan Area from 2015 to 2017. This study is unique in that we found measurement-based evidence of local limiting factors for regional PM formation using spatially dense measurements and satellite data (CrIS surface NH_3).

We found NH_3 concentrations over the NOx-rich SMA to be most highly correlated with $PM_{2.5}$ (R = 0.51) compared to other precursors and PM_{10} (R = 0.32), indicating stronger secondary formation of $PM_{2.5}$, likely due to local ammonia emissions.

These correlation patterns were not repetitive when we applied all nationwide measurements in South Korea (there were no higher correlations found), which implies that the controlling species (e.g., NO_2, SO_2 and NH_3) need to be identified for regional air pollution policy. Doing so would require an assessment of the regional atmospheric chemical environment (e.g., NOx- or NH_3-rich conditions) affected by local emissions and the transboundary transport of pollutants.

Our results show that NH_3 emissions are likely to be a local limiting factor in controlling $PM_{2.5}$ pollution over the NOx-rich SMA. Another recent study with in situ measurements in several parts of SMA also showed that ammonia could be a main controller of fine particle formation [36]. This suggests that the current Korean policy for mitigating NOx emissions from transportation, power plants and industry may not be as effective in some regions (such as the suburbs of the SMA) without an active reduction in local ammonia emissions, mostly from the livestock industry (and in particular from manure). One work showed that a 50% reduction in NH_3 emissions can contribute to a 25% reduction in $PM_{2.5}$ concentrations in winter over Europe [37]. Additionally, unlike other precursors being observed, the relative ammonium concentration has increased recently in Seoul (Han and Kim, 2015). However, a chemical environment with diverse emissions sources (such as the SMA) can be very complex [38]; Seo et al. [7] claimed that a NOx control strategy may be more effective in Seoul considering synergistic nitrate partitioning to the particle phase by wet particles depending on PH conditions. Therefore, more investigations are necessary with measurements of the chemical species of $PM_{2.5}$ to better understand regional NOx or NH_3 limiting conditions, which might vary seasonally. The other limitation of this study is that the relatively low CrIS sensitivity near the surface does not represent direct surface measurements, which implies more local validations of the satellite samples are necessary.

Korean policy for ammonia reduction is still being formulated, as there exist few continuous NH_3-monitoring sites and any reduction targets for NH_3 emissions from the livestock industry will require cooperation between the KMOE and the Ministry of Agriculture, Food and Rural Affairs (MAFRA) [39]. The Korean PM mitigation policy now focuses more on regionally specific pollution controls, through the Basic Plan for Regional Air Quality Management [40].

Korea has very diverse emissions sources in a relatively small spatial area, resulting in numerous complex atmospheric regional chemical environments. Additional comprehensive measurements and analyses of air pollution (including chemically speciated PM precursors) will prove helpful in understanding atmospheric chemical conditions and prioritizing regional PM mitigation policies.

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Data Availability Statement: Derived data supporting the findings of this study are available from the corresponding author on request.

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Conflicts of Interest: The authors declare no conflict of interest.

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